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**Measuring Aerosol Particle Behaviour
due to Human Activity Indoors
for Re-Exposure Evaluation**

A thesis submitted to the National University of Ireland Galway

For the degree of

Doctor of Philosophy

By

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Abstract

The transport and fate of hazardous aerosols, including those of a radiological and biological nature, have been extensively studied both experimentally and theoretically. However, less attention has been given to the subsequent behaviour of the deposited aerosol particles. Deposited aerosol particles can be spread from one surface to another through the transport processes of resuspension and contact transfer; these processes typically involve human activity and can have an important impact on airborne contamination levels and hence, on a person's exposure potential. An experimental study was conducted to quantify the transport process of resuspension of hazardous aerosol particles from clothing surfaces for which limited data currently exist. Four different clothing samples were 'contaminated' with silica particles (3, 5 and 10 μm) labelled with rare earth metals, which were then worn by volunteers who engaged in one of two pre-defined levels of physical activity, within a purpose-built room-sized chamber. Neutron Activation Analysis (NAA), Aerodynamic Particle Sizing (APS) and Scanning Electron Microscopy (SEM) were used to analyse the proportion of the original deposit which was resuspended, as this proportion has a potential for causing re-exposure by the inhalation route. The results show that physical activity can cause up to 67 % of contamination deposited on clothing to be resuspended back into the air. Larger particles are found to be more likely to resuspend; a difference in the general size distribution between deposited and resuspended particles, as well as a shift towards a larger Mass Median Diameter (MMD) within the individual size distributions of resuspended particles (3, 5 and 10 μm) was observed. The NAA data showed an average Resuspended Fraction (RF) of 28 ± 8 %, for initially deposited particles, from the clothing of a person engaged in low physical activity; a corresponding value of 30 ± 7 % was found during high physical activity. The APS data indicated a tenfold increase in the cumulative mass of airborne particles during high physical activity in comparison to that during low physical activity.

It was observed that the contaminated clothing's fibre type had no influence on the levels of particle resuspension from their surface, but that the material's weave pattern (and hence the material's surface texture) significantly influenced the levels of particle resuspension. In addition, following analysis of the airborne mass concentration variation with time during resuspension, the data were found to be separable into two regimes: the first regime showed a high, positive rate of change of airborne particle concentration relative to the second regime, and occurred within the first 1.5 minutes of

the beginning of the resuspension event. The second regime revealed a slower rate of variation in particle concentration.

As a complementary study to the resuspension investigations, experiments were conducted with the aim of quantifying the mass transfer efficiency of deposited particles when various soft and hard surfaces come in contact. The surfaces used were 100 % cotton, synthetic fleece, plastic laminate and brass. Contact transfer efficiencies ranging from 2 to 45 % were observed. Other observations included an increase in the particle mass transferred between surfaces with increased surface roughness. An increase in the applied pressure (from 130 to 9400 Pa) between the two surfaces in contact was seen to lead to contact transfer efficiencies that varied according to pressure, in two distinct pressure regions, with the transition pressure depending upon the surface types in question. The duration of contact between surfaces and the contaminant loading upon them had little effect on the mass transfer efficiencies that were calculated.

The transport of aerosols via the above processes can increase a person's whole body dose following accidental or deliberate airborne releases of hazardous aerosols. The data generated in this work can be used to refine models for radiological exposure assessment. In addition, the results are applicable to biological aerosol transport and infectious disease transmission.

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General Introduction

1 Sources of Hazardous Airborne Particles

There are numerous types of events which could result in the dispersion of hazardous aerosol particles into the air which humans breathe. These hazardous particles are then a potential source of exposure, through routes which will be discussed in section 2. An ‘aerosol’ is defined by Colbeck (1998) as being a dispersion of fine solid particles or liquid droplets in a gas, and for the purposes of this thesis, a ‘hazardous aerosol particle’ will be a general term used to define any aerosol particle which has the potential to harm an exposed person. These aerosols can be of natural or man-made origin and includes aerosols which are radioactive (e.g. carcinogens, ^{137}Cs and ^{131}I from nuclear power plant disasters), biological (e.g. anthrax, moulds and spores, H1N1, tuberculosis) and chemical (e.g. volatile organic compounds, benzene). The origins and potential size distributions of these hazardous aerosol particle types will be discussed in the next sub-sections.

1.1 Radioactive Aerosols

Radioactivity refers to the spontaneous disintegration of an unstable nucleus. The nucleus emits energy in the form of atomic particles (e.g. α or β particles) or as electromagnetic (EM) rays (e.g. gamma or x-rays), and this emitted energy is called radiation. An atom is unstable when it has either too many neutrons or protons. To become stable it can capture an electron, emit a helium nucleus, change a neutron to a proton, or change a proton to a neutron. These are known as decay modes and are individually discussed below.

Alpha decay: The nucleus emits an energetic helium nucleus that consists of two protons and two neutrons. They are large and highly ionising therefore slow down very quickly and deposits a large amount of energy in a short distance i.e. have a high linear energy transfer (LET). They have a short range of approximately a few centimetres in air or a few millimetres in a low density material. Consequently, they ordinarily are not capable of penetrating skin and cause no damage to the tissues below, but are a potential internal hazard through inhalation or ingestion.

Beta decay: Normally a neutron changes to a proton emitting a high-speed electron. This is (β^-) decay and the energetic electron is known as a beta particle. (β^+) decay occurs when a proton changes to a neutron emitting an energetic positron (a positively charged electron). Being composed of charged particles, beta radiation is more strongly ionising than gamma radiation but has a lower LET than an alpha particle. Beta particles can typically travel a few centimetres in plastic or a few millimetres in metal.

Gamma decay: A nucleus changes from a higher energy state to a lower energy state through the emission of quanta of electromagnetic (EM) energy called gamma (γ) rays. Gamma decay often follows alpha or beta decay as the resulting nucleus may not be in its lowest energy state and so must decay to its ground state through the emission of one or more gamma ray photons. Gamma rays have very high energy, typically above 100 keV, and the number of EM photons decays exponentially with distance of travel through a substance such as lead.

1.1.1 Sources of radioactive aerosols

There are many potential sources of radioactive contamination in the environment, including natural and artificial sources. The contribution of naturally occurring radiation to background radiation exposure is far greater than is the case for man-made sources. However, a major accident at a nuclear power plant or an act of terrorism involving large quantities of radioactive material, could result in radioactive material being carried long distances in the atmosphere and affecting populations hundreds of miles away from the source. A countries horticulture and livestock can become contaminated, which exposes the population to contamination by harvesting and ingestion. Inhabitants could also become exposed to the airborne particles through the process of deposition on their hair, skin and clothing. Contaminated residents could then re-expose themselves and others, via the resuspension of these deposited particles.

Human populations are subjected to background radiation every day, from natural and artificial sources. According to the Health Physics Society (accessed in Feb 2012), there are three main sources of natural radiation, the first being the earth, which is the bearer of sources in food and water, for example sodium which is in salt and also sources in building materials. The second source is from space, for example in the form of cosmic

rays; about 9 % of all incoming cosmic ray particles are helium nuclei. The third source is the atmosphere. One major contributor is the radon gas that is released from the earth's crust and subsequently decays into radioactive atoms, which become attached to airborne dust and particulates. Another contribution arises from the radioactive atoms produced in the bombardment of atoms in the upper atmosphere by high-energy cosmic rays.

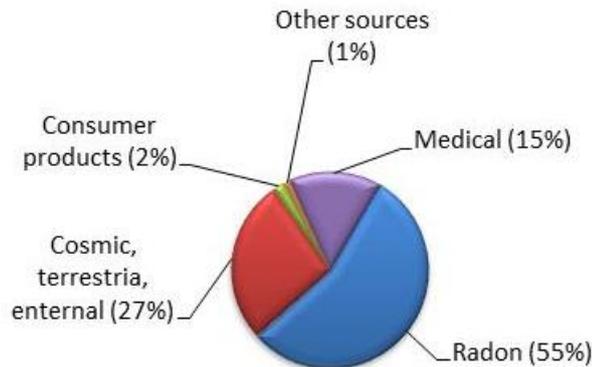


Figure 1. Annual background radiation doses in the US, adapted from NCRP report No. 93, 1987

Artificial radiation accounts for approximately 18 % of the background radiation to which humans are exposed (Radiation Effects Research Foundation). As denoted in figure 1, fifteen percent of that comes from medicine, including x-rays and cancer treatments, and other man made sources are responsible for the remaining 3 %. Some examples include emissions from burning fossil fuels, smoke detectors, nuclear weapons testing, emissions from the normal operation of nuclear power plants or reprocessing plants, and from accidents at these.

The Chernobyl accident which occurred on the 26th of April 1986, is certainly the most publicised, and is widely regarded as the worst accident in the history of nuclear power. In this case, the reactor was undergoing a test to determine how long turbines would spin and supply power to the main circulating pumps, following a loss of mains electrical power supply. The automatic shutdown mechanisms had been disabled by the operator prior to the commencement of the test. The execution of this test resulted in a power surge which caused the fuel rods in the reactor to become overheated and they began to melt. The subsequent explosion was due to chemical reactions, as fuel particles passed into the moderator and coolant, which caused a rapid increase in steam pressure.

The generated steam resulted in the detachment of the reactor lid, the rupturing of the fuel channels and jamming of all the control rods. Intense steam generation then spread throughout the whole core and subsequently blew a hole in the roof. This, plus succeeding explosions of the reactor vessel, along with the resultant fire which burned for ten days, caused vast amounts of radioactive particles to be released into the atmosphere (Anspaugh et al. (1988), estimated that $\sim 100 \times 10^{15}$ Becquerels of ^{137}Cs were released during and subsequent to the accident) and these particles were detected in Scandinavia, and Eastern Europe (Anspaugh et al., 1988, Arvela et al., 1990, Olofsson and Svensson, 1988, Persson et al., 1987). A more recent nuclear power plant accident occurred at the Fukushima Daiichi plant in Japan, on the 11th of March 2011. The accident was as a consequence of the Tōhoku earthquake and ensuing tsunami. The three reactors which were in operation at the time automatically shut down after the earthquake, and emergency generators came online to control the electronics and coolant systems. However, the subsequent tsunami broke the reactors' connection to the power grid, and caused the reactors to begin to overheat, resulting in core meltdowns. The flooding and earthquake damage hindered external assistance and this was compounded by hydrogen gas explosions and the venting of contaminated steam, which released large amounts of radioactive material into the atmosphere (IAEA, 15th of March). The Japanese government estimates that the total amount of radioactivity released into the atmosphere, was approximately one-tenth as much as was released during the Chernobyl disaster (Von Hippel, 2011). Nuclear accidents are rated according to the International Nuclear and Radiological Event Scale (INES), which is logarithmic and ranges from 0 (for an abnormal situation with no safety consequences), to 7 (for a major accident causing widespread contamination and with serious health and environmental effects) (INES user manual). The Japanese Nuclear and Industrial Safety Agency (NISA) temporarily raised the disaster at Fukushima Daiichi to level 7 (IAEA, 12th April). To date, the Fukushima and Chernobyl accidents are the only nuclear accidents with level 7 ratings.

Accidents at nuclear power plants are not a population's only potential source of exposure to radioactive aerosols. Significant amounts of radioactive aerosol could be released into the atmosphere due to transport accidents of nuclear material (no serious accidents to date), during nuclear weapons testing (e.g. Nevada test site, USA), from the release of a nuclear bomb (e.g. Hiroshima and Nagasaki), or due to a terrorist's

deliberate release of radioactive aerosols (e.g. using a radiological dispersion device (RDD) also known as a 'dirty bomb').

An RDD is a standard explosive device packed with radioactive material. This material is dispersed to the air when the bomb explodes. These bombs could vary in size from suitcase-sized devices to devices as large as a truck. Radioactive contamination is left in the immediate area of the explosion site, but contamination may also be spread from the site by wind or by people. A dirty bomb differs from a nuclear bomb in that it primarily contaminates the immediate area surrounding the device but does not include the fission products necessary to create a large blast such as that which occurred in Hiroshima and Nagasaki. The severity of consequence following the detonation of an RDD is dependent on a number of factors including the type of radioactive material used, the size of the radioactive source, the explosion material and mechanism, and the location at which the device is detonated. Although there have been no actual uses of RDD's to date, in 1995, rebels from Chechnya planted, but did not detonate, an RDD device in Moscow's Izmailovo park. This dirty bomb consisted of dynamite and ^{137}Cs , taken from cancer treatment equipment. Information regarding the bomb's location was leaked to reporters, and it was diffused.

Other events that have given rise to radioactive aerosol exposure involve a different exposure mechanism: contact. If a person comes in direct contact with a radioactive source they can become contaminated. The severity of this exposure is dependent on the radioactive source type and age, as well as the length of contact time, among other factors. There have been several published incidences of human exposure due to contact with radioactive sources, both by accidental contact and as a result of malicious intentions. Three accidental incidents (1-3) and three malicious incidents (4-6) are described below, but a more detailed discussion has been published by Mustonen (2009).

1. In Goiana in Brazil in 1985, four people died and many others were seriously injured as a result of the theft of an abandoned teletherapy unit. The source capsule ruptured, exposing the thieves to the ^{137}Cs radioactive source.
2. In Estonia in 1994, one person died and two others were seriously injured following exposure to the ^{137}Cs source they had stolen from a radioactive waste facility. It was three brothers who stole the source and took it home, resulting in extreme radiation exposure to all family members.

3. In Peru in 1999, a construction worker obtained severe radiation burns after accidentally picking up a ^{192}Ir radiography source and placing it in his pocket.
4. In Ecuador in 2002, a criminal gang stole five radioactive sources from a technical company. They demanded, and received, a ransom for the sources, but they only returned three of the five sources.
5. In a Chinese hospital in 2003, a nuclear medicine expert placed ^{192}Ir pellets in his colleague's ceiling as an act of revenge. This resulted in the target person and 74 other hospital staff members suffering from memory loss, fatigue, loss of appetite, headaches, vomiting and bleeding gums.
6. In London in 2006, the former KGB officer Mr. Alexander Litvinenko died of acute radiation effects, just 3 weeks after he suddenly fell ill. Tests established that he had a significant quantity of ^{210}Po in his body, but it could not be determined how this entered his body. Malicious poisoning is regarded as a probable cause and police investigations initially centered on a sushi restaurant in central London and the bar of a London hotel.

Each of the above described incidences, and the previously discussed higher concentration events that have given rise to the dispersion of airborne radioactive aerosols, all resulted in the spread of radioactive particles and hence the contamination of human populations.

1.1.2 Size distribution of radioactive aerosols

According to Andersson et al. (2002), a primary factor governing the impact of radioactive aerosol on the human body is the size of the contaminant particles. Table 1, adapted from table 2 of Dorrian (1997), summarises the size distribution of radioactive aerosol particles in the air, from various sources. Activity Median Aerodynamic Diameter (AMAD) is the median of the distribution of radioactivity or toxicological or biological activity with respect to aerodynamic diameter.

Type of Radioactive Aerosol	Number of measurements	Range of AMAD's (μm)	Median (μm)	Geometric standard deviation [σ_g]
All artificial	125	0.3 – 18	1.5	2.9
Chernobyl fallout	78	0.3 – 4.8	0.64	1.5
Natural ^7Be	56	0.29 – 1.18	0.6	1.9
Resuspended	16	1.025 – 15	6.0	1.9

Table 1. Size distribution of radioactive aerosol (from Dorrian, 1997)

The size distribution of accidentally released radioactive particles from the nuclear working environment depends on the release mechanism, type of fuel used and the temperatures reached (Loyolka, 1983). According to Andersson et al. (2006), with respect to atmospheric transport and exposure, these aerosols can be divided into two groups: a volatile group which includes ^{137}Cs , ^{134}Cs , ^{103}Ru , ^{106}Ru , ^{131}I , ^{132}Te and ^{99}Mo , and a refractory group including ^{89}Sr , ^{90}Sr , ^{141}Ce , ^{144}Ce , ^{95}Zr , ^{140}Ba , ^{95}Nb , ^{240}La and ^{124}Sb . Andersson et al. (2006) also reported the AMAD of radioactive aerosols, outside the immediate vicinity of the Chernobyl nuclear power plant, based on measurements made in various European countries. They found that the AMAD of the volatile radioactive aerosols was of the order of 0.7 μm , in comparison to a significantly higher value of 5 μm for the refractory group.

In the context of nuclear weapons testing at the Nevada test site, when carrying out clean-up and treatment tests, Shinn et al. (1989) detected ^{241}Am and $^{239,240}\text{Pu}$ particles with an AMAD of 5.5 μm . Hirose et al. (1986) measured airborne $^{239,240}\text{P}$ over 14 months and found an AMAD range of 1.6 - 18 μm . At the same time, a range of 3.7 - 8 μm was found for ^{90}Sr . Uranium mills yield an AMAD range of 0.72 - 20 μm and have a median value of 6.8 μm as reported by Dorrian (1997).

1.2 Bioaerosols

Bioaerosols are formed by suspension of particles of biological origin in the air (Lacey and Dutkiewicz, 1994). They include: viruses (e.g. H1N1), living organisms (e.g. bacterial and fungi), and parts of products of organisms (e.g. fungal spores, pollen and animal allergens). Bioaerosols are common to both indoor and outdoor environments but are usually found at low concentrations. Natural background concentrations of bacterial, pollen and fungal spores are typically 0 – 10,000 units m⁻³ (Hinds, 1999). The most commonly found bioaerosols are bacteria and fungal spores.

1.2.1 Sources of bioaerosols

Bioaerosols arise from plants, animals, humans, soil and water and can be divided into two main groups: *viable* and *nonviable*. Viable bioaerosols include any living organisms; they can be identified and quantitated by growing individual organisms with suitable nutrients into visible clusters or colonies, and they show a decay in biological activity with time that depends on the relative humidity, oxygen content, and the concentrations of trace gases in the air (Hinds, 1999). Nonviable bioaerosols are dead organisms, and include pollen, animal dander, and insect excreta. Some specific categories of bioaerosols are discussed below.

Bacteria are free-living single-celled organisms with an average size of approximately 0.5 µm (Cherrie et al., 2010). They are usually spherical (cocci) or rod shaped (bacilli) but may occur as clusters or chains. Bacteria tend to colonize water or soil and are subsequently released as aerosols when the water or soil is disturbed. Some bacteria release a hardy, dormant version of bacteria called ‘endospores’ (Hinds, 1999). These are insoluble and toxic. The primary sources of indoor bacterial aerosols are water-containing devices, such as humidifiers and cooling towers, and stored organic material. Examples of bacterial pathogens and their associated diseases are: *Bacillus anthracis* - anthrax, *Yersinia pestis* - plague, and *Francisella tularensis* - Tularemia.

Fungi may occur as single-celled organisms (e.g. yeast), but are more commonly found as microscopic, multicellular, branching structures called ‘hyphae’ (e.g. mold and mildew) (Hinds, 1999). Fungi are most frequently found in indoor damp locations, in soil (e.g. *Basidiomycetes*), in stored organic material (e.g. *Aspergillus* and *Penicillium* species), and on decaying vegetation. They spread by releasing spores into the air. They

also excrete enzymes and toxins into the air and the combination of excreted materials is called ‘mycotoxins’ (Ledford, 1994). These metabolites have been identified as being toxic to humans, for example, aflatoxins, which are carcinogenic, are mycotoxins produced by many species of *Aspergillus* (Cherrie et al, 2010).

Viruses (the smallest infectious agents) are intracellular parasites, consisting of genetic materials surrounded by a protein coat, and can reproduce only inside other living cells (Cherrie et al., 2010). Alone, a virus is usually sub-micron in size, but airborne viruses are typically found as part of a droplet nuclei or attached to other airborne particles and therefore vary in size. Viruses are transmitted through direct contact, through contaminated food or water, or through exhalation. Examples of viral pathogens and their associated diseases are *Variola major* - smallpox, *SARS associated corona virus* - severe acute respiratory syndrome (SARS), and *Ebola virus* - ebola hemorrhagic fever.

Pollen grains are near-spherical particles produced by plants. Pollen is often aerosolised via wind suspending the pollen from the end of floral anthers, and it is therefore highly seasonal and dependent on wind and weather.

The above paragraphs describe the types, and general environmental sources of bioaerosols. However, exposure to bioaerosols may also occur by malicious means or terrorist activity. Bioterrorism is defined by Brachman (2002) as ‘the use, or threatened use, of biologic agents against a person, group, or larger population to create fear or illnesses for purposes of intimidation, gaining an advantage, interruption of normal activities, or ideological objectives’. This will typically involve a release of much higher concentrations of the particular bioaerosol than are generally found in the natural environment.

Prior to the relatively modern phenomenon of intentional anthrax releases, humans contracted the human anthrax disease due to contact with animals, or animal products, which were contaminated with *Bacillus anthracis* spores. Since the mid-20th century, there have been much stricter regulations on industrial hygiene practices and the importation of animal products, and thus outbreaks of this type are now relatively uncommon. The first confirmed outbreak of intentional anthrax release in the United States, which is also the largest bioterrorist incident relating to anthrax contamination to date, occurred between October 4 and November 2, 2001, in the District of Columbia, Florida, New Jersey, and New York (Jernigan, 2001). According to the Centers for

Disease Control and Prevention (CDC), there were 10 confirmed cases of inhalational anthrax (which has a typical mortality rate approaching 100 %, Dixon et al., 1999) and 12 confirmed or suspected cases of cutaneous anthrax (the most common form and usually curable, Dixon et al., 1999). The contamination resulted from the intentional delivery of *B. anthracis* spores through mailed letters or packages. In the case of this outbreak, 60 % of patients survived due to a combination of early detection, multidrug antimicrobial therapy (begun during the initial phase of the illness), and aggressive supportive care (Jernigan, 2001). This survival rate is much higher than was the case for any other incidences of anthrax inhalation. For example, in the city of Sverdlovsk in the former Soviet Union in 1979, only one in seven of those who contracted inhalation anthrax survived (an 86 % mortality rate), following the accidental release of an aerosol containing *B. anthracis* spores from a military biological facility, where the aerosol was produced for the purpose of warfare (Brachman, 2002).

Another type of incident involving large scale bioaerosol contamination, is the global epidemic of antimicrobial resistance (AMR) and healthcare-associated infection (HAI) contraction. These are among the most serious public health problems in the European Union (EU); each year, approximately 4 million patients acquire a healthcare-associated infection and approximately 37,000 of them die as a direct result of the infection (European Centre for Disease Prevention and Control (ECDC), Annual Report of the Director, 2010). One highly prevalent infection, acquired in hospitals worldwide, is methicillin-resistant *Staphylococcus aureus* (MRSA). The European Antimicrobial Resistance Surveillance System (EARSS)'s annual report of 2002 stated that between 1999 and 2002, there was a significant increase of MRSA rates in Austria, Belgium, Germany, and the United Kingdom. Germany and Austria witnessed the quickest expansion of MRSA rates (proportional to the country's population): from 8 % to 19 % and from 5 % to 11 %, respectively, while the United Kingdom and Ireland saw more of a stabilisation of the fast rise that characterised the increase of MRSA throughout the 1990's, and they had maintained levels below 45 % in 2001 and 2002. Between 1989 and 1997, at the University Hospitals of Geneva in Switzerland, 1771 people contracted MRSA (Harbarth et al., 2000). Measures to control MRSA outbreaks have concentrated on transmission of the organism and prospective screening for carriage, in combination with general infection control measures such as patient isolation, use of barrier precautions, and environmental decontamination, but despite these measures occurrences continue to increase (Monnet, 2004). Since antimicrobial-resistant

microorganisms (such as MRSA) are difficult to treat medically, infections due to these microorganisms result in prolonged illness/stays in hospitals and an increased risk of death (ECDC, 2010).

Legionellosis is a potentially fatal infectious disease caused principally by the organism *Legionella pneumophila*. The disease takes two forms, the most severe form of which is, Legionnaires' disease, an uncommon form of pneumonia. The second form is called Pontiac fever, and is caused by the same bacteria but produces a milder respiratory illness without pneumonia that resembles acute influenza. In the case of Legionnaires' disease, there is a fatality rate of 11 % in those cases with a known outcome, and about 5,000 to 6,000 cases are reported in the EU each year (ECDC, 2010). According to the CDC, between 8,000 and 18,000 people are treated for Legionnaire's disease each year in the United States and the fatality rate is between 5 % and 30 %. Most cases can be successfully treated with antibiotics. As with anthrax, the infection cannot be spread by human-to-human transmission but is normally as a result of inhalation of environmentally sourced (e.g. infected water) aerosols containing *Legionella* bacteria. The outbreak with the highest number of deaths to date (34 deaths from 221 cases) occurred in Philadelphia, Pennsylvania in 1976. However the outbreak with the highest mortality rate - 33.3 % - occurred more recently in St Peter's University Hospital, in New Brunswick, New Jersey, in 2008. Here two of the six people infected died as a result of the chlorination levels in the hospital's water system dropping below effective levels.

Influenza is a commonly spread viral infection that affects mainly the nose, throat, bronchi and, occasionally, lungs. According to the World Health Organization (WHO), infection usually lasts for about a week, and is characterized by sudden onset of high fever, aching muscles, headache and severe malaise, non-productive cough, sore throat and rhinitis. According to the ECDC Annual Report of the Director 2010, each winter, epidemics of seasonal influenza cause up to 40,000 premature deaths in the EU and EEA/EFTA countries. The report states that while there are no figures for the total morbidity each year, but the estimates are that influenza affects around 5 - 10 % of the population each season, with higher rates in younger people. Unlike legionellosis, MRSA and anthrax, this virus can be easily transmitted from person to person via droplets and small particles produced when infected people cough or sneeze. Influenza pandemics occur sporadically, with the emergence of new human infections such as

avian influenza A (H5N1) and Severe Acute Respiratory Syndrome (SARS), both of which had profound effects on the international health community. The first human outbreak of H5N1 was detected in 1997 in Hong Kong. Although the virus proved difficult to transmit to humans, case fatality was high and as of February 2011, about 500 laboratory-confirmed human cases, from 15 countries, had been reported to WHO, about 60% of which were fatal (WHO, report by Director General, 2011). The first cases of SARS appeared in November 2002 in Guangdong Province, China. By the official end of the outbreak on July 5th 2003, WHO reported 8,098 cases and 774 deaths, from 26 countries. In contrast to H5N1, SARS was readily transmitted via the person-to-person route, most likely through respiratory secretions, as was the most recent global influenza pandemic, the swine influenza (H1N1) outbreak which began in early 2009. By August 2010, when WHO announced the transition from pandemic to post-pandemic period, about 18,500 laboratory-confirmed deaths from pandemic influenza A (H1N1) 2009 had been recorded (WHO, report by Director General, 2011).

Each of the above described large scale sources of bioaerosol contamination to humans, provides justification for the necessity of fully understanding the mechanisms by which aerosols are spread in the environment, in order to design effective countermeasures.

1.2.2 Size distribution of bioaerosols

As particle size determines the fate of particles on/in the human body which thus affects a person's exposure risk, it is necessary to fully understand the size distribution of aerosol particles when determining secondary exposure.

As the term 'bioaerosols' covers a wide range of aerosol types, the size range of bioaerosols is correspondingly large. As discussed in section 1.2.1, bioaerosol types include; bacteria, fungi, viruses and pollen, each with a size distribution as summarised in table 2.

Bioaerosol type (specific sub-division)	Size range (μm)
Bacteria Endospores	0.3 – 10 0.5 – 3.0
Fungi Fungal spores [#] Hyphae Mycotoxins	0.5 – 10 x 10⁸ 2.0 – 50 2.0 – 4.5 0.5 – 30
Viruses	0.005– 0.3
Pollen[#] Most common sizes	5.0 – 200 25 – 50

Table 2. Size distributions of bioaerosols (data predominantly from Hinds, 1999, [#]from Cox and Wathes, 1995)

The environment in which bioaerosols are found, i.e. whether indoors or outdoors, determines the concentration and type of bioaerosol to which one is exposed. Exposure is likely to be higher in the indoor micro-environment than outdoors due to the amount of time people spend there (Colbeck and Nasir, 2010). Simoni et al. (2003) estimated that a minimum of 80 % of people's lives are spent indoors and therefore it can be concluded that a knowledge of the type, concentration and size distribution of bioaerosols indoors is of major importance. Nasir and Colbeck (2010) investigated the concentration and size distribution of bacteria and fungi in 15 houses which were categorized into three types: type I - a single room in shared accommodation, type II - a single bedroom flat in a three story building, and type III - two or three bedroom houses. Their findings, in terms of bioaerosol size distribution, are summarised in table 3.

Housing type	Size distributions (in μm) of maximum levels of culturable:		Average geometric mean diameters (in μm) of:		Geometric Standard Deviation	
	Bacteria	Fungai	Bacteria	Fungai	Bacteria	Fungai
Type I	3.3 – 4.7	4.7 - 7.0	4.70	4.50	1.50	2.90
Type II	3.3 – 4.7	2.1 – 3.3	3.89	3.57	2.30	3.60
Type III	3.3 – 4.7	2.1 – 3.3	1.96	3.92	2.50	1.38

*Table 3. Size distributions of bioaerosols in homes
(data from Nasir and Colbeck, 2010)*

The size distributions of some specific viruses, bacteria and fungi that are responsible for high risk exposure and large scale human contamination are discussed in the succeeding paragraph.

Infectious bioaerosols are typically between 0.1 and 10 μm and the majority of viruses and bacteria that cause respiratory diseases in humans are generally associated with bioaerosols with diameters greater than 5 μm (The Institut de recherche Robert-Sauvé en santé et en sécurité du travail (IRSST), 2007). Viruses with the potential of being released for the purposes of bioterrorism, such as measles, varicella and smallpox, are typically 0.3 μm in size. Anthrax spores are approximately 1 to 2 μm (Dixon et al., 1999) in diameter, a size that is optimal for inhalation and deposition in the alveolar spaces, as will be discussed in section 2. *H. Capsulatum* is a species of pathogenic fungi, typically > 1 μm in size, which causes histoplasmosis - an infectious disease in humans. Influenza pandemics (e.g. avian H5N1 and swine H1N1) can be transmitted by droplets, typically > 5 μm , or in aerosol form, typically \leq 5 μm (IRSST, 2007).

2 Exposure to Hazardous Airborne Particles

It is important to determine the route by which a human becomes exposed, as the impact of hazardous aerosol particles on health is determined by their exposure route. The particle size will have influence on a person's exposure route and severity of exposure. There are many routes for human exposure to hazardous particles including ingestion, inhalation and absorption through the skin, and the size of the particles associated with these exposure routes varies. The three main exposure routes to the human body are discussed below.

2.1 Ingestion

If animals or crops become contaminated through either the atmosphere or water supply and are then ingested by a human, they will become contaminated. Also human's drinking water supplies may contain hazardous aerosol particles. Furthermore, transfer of contamination via contact of hands or food with a contaminated surface can lead to ingestion of hazardous particles. There are many examples of this in the food sector; for example, Dawson et al. (2007) measured the transfer of *Salmonella* Typhimurium from wood, tile or carpet to bologna (sausage) and bread. They concluded that *Salmonella* Typhimurium can survive for up to 4 weeks on dry surfaces, in high-enough populations to be transferred to foods and that this could increase the risk of foodborne illnesses through ingestion.

In the context of radioactive contamination via ingestion, Olofsson and Svensson (1988) examined the pathways of ^{131}I , ^{134}Cs and ^{137}Cs from the Chernobyl fallout to man in Västerbotten, Sweden. They concluded that the most important sources of Cs intake in man were from ingestion of lake fish, elk (European moose) and reindeer. According to Phillips (2002), fourteen years after the Chernobyl nuclear accident, intake of radionuclides is almost exclusively as a result of drinking contaminated water and milk, and eating foods grown in contaminated soil. It was not until 10 years after the Chernobyl accident that deep artesian wells were dug in the city of Kiev (the capital city of the Ukraine, located 72 miles from the reactor), as sources of safe drinking water for residents.

2.2 Inhalation

Hazardous aerosol particles which are suspended in the atmosphere can be inhaled into the human lungs, and particle size is a major factor in inhalability. Only 50 % of aerosols larger than 50 μm will be inhaled (Kennedy and Hinds, 2002); some of these hazardous aerosols are re-exhaled but the remaining ones are immediately absorbed by the lung tissue and can have adverse effects on lung tissue, as will be discussed in section 3. Following inhalation, particles which are not exhaled will deposit onto the tissues of the lung due to a number of deposition mechanisms or may be exhaled. The three core deposition loss mechanisms are inertial impaction, sedimentation, and diffusion. Inertial impaction occurs when a particle's momentum is sufficient for it to deviate from streamline fluid flow and is the primary mode of particle deposition in the respiratory region (Mark, 1998). It is most prominent for particles $> 1 \mu\text{m}$ and for high velocity particles. Sedimentation is most prominent for low velocity particles and its likelihood increases with increasing particle size. Diffusion occurs due to particles undergoing random Brownian motion (which is independent of any convection associated with the air itself, Mark, 1998) and colliding with a surface of the respiratory tract. It is the primary deposition mechanism for particles $< 0.1 \mu\text{m}$ in size (Byrne, 2009). The likelihood of these deposition loss mechanisms occurring is dependent on: fluid dynamics, airway geometries and the particle's physical characteristics (Byrne, 2009).

Hinds (1999) states that for the purposes of examining particle deposition in the respiratory system, it can be divided into three separate regions, each distinctly differing in structure, airflow patterns, function, retention time and sensitivity to deposited particles. The first region is the *head airways region* (also called the extrathoracic or nasopharyngeal region) and is comprised of the nose, mouth, pharynx and larynx. The second region is the *lung airways or tracheobronchial region*. This includes the airways from the trachea to the terminal bronchioles. The third region is the *pulmonary or alveolar region* which includes the respiratory bronchioles, alveoli. Particles of almost any size can be inhaled but the depth to which particles can penetrate the lungs is highly dependent on their aerodynamic diameter. Particles greater than 10 μm in diameter cannot reach the alveolar region of the lungs and particles greater than 30 μm will not penetrate the respiratory region past the larynx (Cherrie et al., 2010).

Throughout history, there are extensive examples of human contamination via inhalation. Healthcare-associated infections such as MRSA, and viruses such as influenza (SARS and N1H1), are all primarily contracted via inhalation. Inhalation of *Bacillus anthracis* spores causes human anthrax disease, which was prevalent in the textile industry in the mid-1800's. Due to the frequency of incidence of infection in mill workers exposed to imported animal fibres contaminated with *B. anthracis* spores, inhalational anthrax related to the textile industry became known as woolsorters' disease in England and ragpickers' disease in Germany and Austria (Jernigan, 2001).

A consistently active, global research topic is the study of carcinogens – which is any substance, radionuclide, or radiation that is directly involved in causing cancer. Common examples of carcinogens are inhaled asbestos and tobacco smoke. The International Agency for Research on Cancer (IARC) categorises all agents, groups of agents, and mixtures, which are carcinogenic, into five groups. Group 1 (107 agents) are characterised as being 'carcinogenic to humans' and include: arsenic and arsenic compounds, asbestos, benzene, beryllium and beryllium compounds, hepatitis B virus (HBV) and hepatitis C virus (HCV), and tobacco smoke. Group 2A (61 agents) are characterised as 'probably carcinogenic to humans' and include: acrylamide, benzidine based dyes, cisplatin, diesel engine exhaust, dimethyl sulphate, and formaldehyde. Group 2B (269 agents) are characterised as 'possibly carcinogenic to humans' and include: glasswool, lead and inorganic lead compounds, and methylene chloride. Group 3 (508 agents) are characterised as being 'unclassifiable as to its carcinogenicity to humans', and group 4 (1 agent) characterised as 'probably not carcinogenic to humans'. Kauppinen et al. (2000) stated that at least 22 million workers in the EU were exposed to IARC group 1 carcinogens and some of the most common exposures included: environmental tobacco smoke (7.5 million workers exposed at least 75 % of working time), crystalline silica (3.2 million exposed), diesel exhaust (3.0 million), and wood dust (2.6 million). Skwarzec et al. (2001) stated that the main sources of ^{210}Po and ^{210}Pb intake of smokers in Poland, was from cigarettes (as opposed to food) and its principal pathway was absorption through the respiratory system.

2.3 Dermal

The skin is the largest organ in the human body and serves as a physical barrier between the body and the environment. It can be divided into two distinct layers. The first is the *Epidermis* which is the thin outer section and comprises of several layers, the outermost of which is called the stratum corneum and the deepest is the basal layer. Beneath this is the *Dermis* which is the thickest section and is a complex structure of blood vessels, nerves, lymphatic vessels, hair follicles and sebaceous glands (Charles, 2004). Aerosol particles may deposit onto the skin's surface from the surrounding contaminated air. If they are of appropriate size, they can penetrate the skin's surface or could be absorbed through a lesion in the skin. Another process for dermal contamination is through contact transfer, as skin comes in contact with hazardous aerosol particles which have deposited to a surface. For hazardous particles deposited onto skin; particles greater than 10 μm cannot penetrate the skin, particles of 3 – 10 μm may reach deeper layers of the skin through the hair follicles but particles less than 3 μm can diffuse through the stratum corneum to be incorporated into the body's blood supply (Shekunov et al., 2007 and Williams, 2003). These smaller particles ($< 3 \mu\text{m}$) are therefore of high exposure risk as they cannot be safely removed if they have penetrated through the skin. Particles larger than 1 μm will remain on the skin until they are removed or resuspended (Andersson et al., 2004); this can lead to re-exposure through any of the above mentioned pathways.

There are numerous published examples of human contamination via dermal exposure. In the context of radiological contamination, incidences of dermal exposure following the Chernobyl accident include that of two women who were vacationing in Kiev shortly after the accident and who received light radioactive contamination in and on their bodies, and on their clothing (Beentjes et al., 1988). There are also published papers on the dermal contamination of workers and fire fighters resulting directly from the Chernobyl accident. Barbanova and Osanov (1990) discuss the conditions of exposure of 56 victims who suffered radiation lesions in the skin. The skin of six of those patients (excluding the 29 plant operators, six firemen, and 15 persons exposed to distant β - γ sources only) was exposed to fallout particles, which formed thin radioactive sources on the skin surface. Those victims were located at various distances from the plant but were all within the area over which the radioactive plume travelled. The most exposed parts of the body were the head (face), neck, shoulders and hands, in

that order. Dermal contamination via contact transfer has been examined in the medical industry. Reed (1975) studied the transmission of Rhinovirus colds through indirect skin contact and found that the virus could be spread from an infected person to another person via dermal contact with intermediate objects.

Humans can become exposed to hazardous particles through any of the above routes however; secondary exposure may also occur via two primary aerosol particle transport processes, resuspension and contact transfer. Resuspension occurs when initially deposited aerosol particles are re-entrained back into the atmosphere when the surface on which the particles are deposited is disturbed (e.g. a human resuspending aerosol particles by walking on a contaminated floor). Contact transfer occurs when particles are transferred between surfaces via contact of the surfaces (e.g. a hand collecting particles by touching a contaminated surface and subsequently touching a clean surface, such as another person's hand). These aerosol particles are then available once again, to re-expose humans via any of the above determined exposure routes, and these secondary exposure pathways provide a direct link between exposure routes. Therefore, airborne or surface concentrations should not be viewed as independent variables but rather as variables with a co-dependency; this will be discussed further in section 4.

3 Health Effects

3.1 Radioactive Aerosols

It is important to note that a person exposed to radiation is not necessarily contaminated with radioactive material. Exposure means that radioactive waves or particles have penetrated the body, while contamination infers that radioactive material must be on or inside the body. External contamination occurs when radioactive material comes into contact with the person's skin, hair or clothing. This can lead to internal contamination which occurs when radioactive material is inhaled, ingested or absorbed through the skin (as discussed in section 2).

In the context of radiation exposure there are a few terms used to describe the associated radiological risk to humans. *Dose* (D) is a measure of the amount of radiation a piece of material (e.g. skin) has absorbed. It is the fundamental dosimetric quantity used and is

measured in units of *Grays* (Gy) (one joule per kilogram of the material). However, dose it does not take into account the type of radiation or how it interacts with matter. A more comprehensive measure is the *Equivalent dose* (H_T), which takes the type of radiation into account and has the unit *Sievert* (Sv). It is the average absorbed dose (in Gy) modified by the radiation weighting factor w_R . The most comprehensive measure is the *Effective Dose* (E), which takes into account the type of radiation and the tissue or organ it affects, also in units of Sieverts (Sv). It is the average equivalent dose modified by the tissue weighting factor w_T .

The factors which determine the dose received by the body from material deposited or adsorbed on its surfaces include the area of contaminated surface, the quantity of contaminating material deposited and the length of time the contamination is on the body i.e. exposure time (Byrne, 2009). Radioactive contamination of the skin is a complex topic due to the number of variables involved. The main biological effects which can be induced by ionising radiation are divided into three categories by Charles (2004): early and late deterministic effects, and cancer induction. Several types of early deterministic effects can be produced by penetrating radiations, such as high-energy beta radiations or X radiation and gamma radiation. These categories of severe irradiation-related skin damage are described by Charles (2004) and summarised below.

Erythema is a skin-reddening effect produced by acute radiation doses of at least several grays to the superficial blood vessels of the upper dermis. *Moist desquamation* is the result of death of the epidermal basal cells and damage to the superficial network of blood vessels. Healing can be rapidly achieved, providing that epithelial cells around the base of the hair follicle survive to provide a source of cell repopulation. *Ulceration and Necrosis* are the result of deep dermal structure damage and occurs 2 – 10 weeks following exposure. *Dermal atrophy and damage to the deep vasculature* are effects that become apparent on a timescale of months or years and are hence the main late effects following an acute exposure.

External contamination can also occur due to exposure to ‘hot particles’ which are “physically small radioactive sources with linear dimensions up to approximately one mm” (Byrne, 2009). Charles et al. (2000) reviewed the origins, physical and radiological characteristics, biological effects, and international dose limits for non-respirable, radioactive hot particles which present potential hazards to the skin. He

stated that these particles range in size from $\sim 10 \mu\text{m}$ up to $\sim 2 \text{mm}$ and that the superficial skin dose is dominated by beta particle emission. The National Council on Radiation Protection and Measurements (NCRP) concluded in Report 106, that for beta-emitting hot particles on the skin, the effect to be controlled is “acute deep ulceration”.

Harmful radioactive particles can also contaminate the human body through the respiratory system when inhaled. The lung is considered to be the most critical organ for cancer induction due to exposure to both naturally occurring and man-enhanced ionising radiation in the environment (Burkart, 1989). Following inhalation, particles will deposit onto the tissues of the lung (as discussed in section 2), consequently causing internal contamination. The severity of the dose received is primarily dependent on the particle type, shape and size, as well as the deposited location within the respiratory tract. Following radiation inhalation, the critical injuries that eventually lead to impaired ventilation and diffusion capacity are related to the total dose, its fractionation, and to the volume of the lung irradiated (Coggle et al., 1986). The 1986 publication by Coggle et al. describes the effects of radiation in the lung as well as using experimental data to describe the significant early and late reactions of the lung to radiation exposure. Their conclusions are summarised below.

At very high doses (tens of Gray), radiation can cause the cessation of metabolism and cellular disintegration, a type of death known as interphase death. Radiation causes acute effects (within days or weeks) in rapidly proliferating tissues and delayed effects (months or years) in slowly or non-proliferating tissues. In terms of acute effects, the lung's response can be divided into two syndromes: (a) *radiation pneumonitis*, which develops within 6 months after exposure to doses $\geq 8 \text{Gy}$ of X- or γ - rays and (b) *radiation fibrosis*, which is a delayed or late reaction that develops from about 6 months to years after exposure. Radiation pneumonitis is generally regarded as an inflammatory reaction that progresses to a chronic fibrotic reaction. Deaths can occur in both the acute and the late phase. Lung cancer can also be induced following exposure of the lungs to radiation. For example, uranium miners received their irradiation primarily from the inhalation of airborne alpha radiation. Uranium mines emit radon and thorium gases and diffused radon-222 gas rapidly decays (half-life 3.8 days) to yield radon daughters, two of which are isotopes of polonium. These atoms quickly attach to dust and water vapour to produce respirable aerosols. Lung cancer is an area of extensive research due to the high mortality rate associated with the disease.

3.2 Bioaerosols

The principle route of entry of bioaerosols into the body is via inhalation. For some microorganisms there is a possibility of skin infection (e.g. cutaneous anthrax) or the transfer of the material into the body by ingestion from hand-to-mouth contacts (e.g. Hepatitis A virus) (Cherrie et al., 2010).

According to the IRSST (2007), two classes of biological hazards exist, based on the characteristics of the microorganisms making up a bioaerosol. They are:

1. Infectious hazards (e.g., viruses, pathogenic bacteria). Infectious bioaerosols must be living to cause infections, which are defined as being the result of the penetration and development in a living being of microorganisms that can cause lesions by multiplying, and eventually by secreting toxins, or by spreading through the bloodstream.
2. Non-infectious hazards (e.g., non-pathogenic bacteria and molds). Non-infectious bioaerosols include microorganisms found in the environment in general, which, even when dead, can produce immunological or toxic reactions when inhaled. Molds are an example of these microorganisms.

The diseases associated with bioaerosol exposure have been categorised by Douwes et al. (2003) as infectious diseases, respiratory diseases and cancer. Bacteria which cause human disease are called 'human pathogens'. Infections from pathogenic viruses, bacteria and fungi can occur, but the more typically encountered symptoms, as listed by Lacey and Dutkiewicz (1994), include mucous membrane irritation, bronchitis and obstructive pulmonary disease, allergic rhinitis and asthma, allergic alveolitis (granulomatous pneumonitis) or organic dust toxic syndrome (inhalation fever or toxic pneumonitis). Hinds (1999) discusses some of the more serious health effects caused by bioaerosol exposure, and they include infectious disease, sensitisation reactions, and irritations.

Infectious disease: Infectious diseases are caused by inhalation of (i) aerosolised bacteria, e.g., tuberculosis, legionellosis and inhalation anthrax, or (ii) fungi or spores, e.g. histoplasmosis and aspergillosis, or (iii) viruses, e.g. colds, flu, chicken pox and measles. Approximately 10 % of the entire burden of disease in European countries (which include new and old member states of the European Union) can be attributed to infectious diseases (EARSS, 2002).

Most fungi are associated with *sensitisation reactions* e.g. asthma, but pollen can also cause allergic diseases such as hay fever. Curtis et al. (2004) reviewed 17 studies and concluded that 6 – 10 % of the general population and 15 – 50 % of atopics had immediate skin sensitivity to fungi. Asthma is one of the most common chronic diseases in the world and it is estimated that up to 300 million people suffer from asthma, and that asthma accounts for about 1 in every 250 deaths worldwide (Masoli et al., 2004). Woolcock (1986) revealed that New Zealand had the highest death rates and prevalence of asthma worldwide, and that approximately 0.8 % of the Australian population is at risk of death from asthma and 4 % are at risk of developing severe and irreversible lung disease.

Reactions to toxins or irritants such as endotoxins (components of cell walls of certain bacteria) and mycotoxins from fungi can occur. Some common mycotoxins, as listed by Curtis et al. (2004), include: Aflatoxins (which are very potent carcinogens and hepatotoxins, produced by some *Aspergillus* species); Ochratoxins (which are nephrotoxic and carcinogenic, and are produced by some *Aspergillus* and *Penicillium* species); Sterigmatocystin (which is immunosuppressive and a liver carcinogen, and is produced by *Aspergillus* species), and Trichothecenes, which is produced primarily by *Stachybotrys* and *Fusarium* species, and have been reported to inhibit protein synthesis and cause haemorrhage and vomiting.

According to Portnoy et al. (2005) reducing fungal exposure can reasonably be expected to improve health. Removal of moisture from the indoor environment and proper maintenance of air filters can aid in prevention and elimination of fungi from the home environment. However, indoor exposure to *Aspergillus* and other fungi, can lead to life-threatening systemic infections in immunocompromised patients (Curtis et al., 2004). Several diseases may arise from inhalation of fungal spores in the course of handling decaying matter, faeces, compost or soil. These diseases include aspergillosis, histoplasmosis, blastomycosis, coccidioidomycosis and adiaspiromycosis (Douwes, 2003). The health problems associated with fungi exposure include; fatigue, rhinitis, memory loss and other neuropsychiatric problems, respiratory problems, fibromyalgia, irritable bowel syndrome, vasculitis and angioedema. Denning (1996) concluded that for cerebral, pulmonary, and sinus aspergillosis, case-fatality rates (CFRs) were 99%, 86%, and 66%, respectively and that no untreated patient survived.

Figure 1 in Dixon et al. (1999) depicts the pathophysiology of anthrax to the human body. The symptoms of inhalational anthrax are described by Brachman (1980), which consists of two phases. The first phase includes symptoms of malaise, fatigue, fever, myalgias, and nonproductive cough. This typically lasts one to four days, following which there can be improvement in the patients' clinical condition. The second phase consists of respiratory distress, cyanosis, and diaphoresis. Consciousness is typically maintained until death, but with meningeal involvement there may be disorientation, coma, and meningismus. Death, in one to two days, almost always follows the onset of the second phase.

4 Transport Processes for Contaminant Re-distribution

There are many transport processes by which contaminants can be re-distributed in an indoor environment. These processes are described by Schneider et al. (1999) and include *emission* of particles from the primary source, *deposition* of particles from the air to a surface (e.g. clothing, skin, indoor surface), *resuspension* or *evaporation* from a surface to the air, and *transfer* or *removal* from clothing to skin or vice versa, or from skin to a surface or vice versa, via contact of the surfaces. These transport processes can change the route by which a person can become exposed to hazardous aerosol particles.

A person's inhalation exposure is directly proportional to the concentration of particles in the air (Fogh et al., 1997) and dermal exposure is directly proportional to the concentration of particles deposited on the surface of their skin, as well as the concentration of particles deposited to other indoor surfaces which may be transferred to the skin via contact. However there is a co-dependency between surface (whether skin or another surface) and airborne particle concentrations. It has been well documented that airborne particles will be transported in the atmosphere and eventually deposit onto a surface. While initially airborne they are a potential inhalation exposure risk, then, following deposition to a surface, they are a potential dermal exposure risk (either through direct deposition to the skin, or deposition to a surface which then comes in contact with skin). The subsequent disturbance of the surface can then cause the deposited particles to become resuspended into the air and once more become a potential inhalation exposure risk for humans. Therefore the particle transport process of resuspension will change a person's exposure from dermal to inhalation, and the

transport process of contact transfer can increase dermal exposure and also lead to ingestion exposure. The specific means by which the processes of resuspension and contact transfer can change a person's exposure route will be discussed in the following sub-sections.

4.1 Resuspension

Deposited aerosol particles can be resuspended into the air by a number of different means, which will be elaborated upon below.

Particles deposited onto flooring or onto furnishings inside a building, may be resuspended simply due to air currents lifting the particles from the surface. Likewise, particles on outdoor surfaces, such as grass swards or roads, can be entrained to the air as a result of the wind. Nicholson (1993) used a wind tunnel to investigate the resuspension of 4, 10, 18 and 22 μm sized particles, from grass and concrete surfaces, as a function of wind speed and time. Nicholson concluded that there was an increase in resuspension rate with increasing wind speeds, and that this relationship was most pronounced for the larger sized particles. However, a more common process by which particle resuspension can occur involves the agitation or disturbance of the surface on which the particles are residing. The physical movement of contaminated surfaces as a mechanism for resuspension has been examined by many researchers. Andersson et al. (2004) studied the variation in levels of resuspension due to the dropping of a weight onto various contaminated surfaces. It was concluded that, in comparison to wood, plastic provided the least resistance to resuspension due to its smoothness. However, the dropping of a weight, or an increase in air velocities indoors, can most probably be attributed to human movement. Hence a common source of surface disturbance, and hence particle resuspension, is typically human physical activity.

Human physical activity is a broad term encompassing a wide range of movement types or physical activities ranging from small movements which simply cause changes in air currents, to large scale movements such as jumping or dancing which can cause a surface to move violently. In published research to date, two main surface types have been examined in relation to particle resuspension due to human physical activity. These surfaces include flooring and human body surfaces (skin, hair and clothing).

4.1.1 Resuspension from flooring

A review of the literature has revealed that the majority of currently published research on resuspension focuses on resuspension from flooring of various types. Ferro (2004) examined the effect of low and high physical activity – walking or dancing – on particle source strengths from various floor surfaces. Dancing on a rug resulted in PM_{2.5} and PM₅ source strengths three times larger than walking on the same rug. Vacuuming resulted in slightly higher PM₅ source strengths than simply walking and sitting on furniture. Long et al. (2000) estimated that dusting and vigorous walking contributed 23 and 12 µg m⁻³, respectively, to indoor PM_{2.5} concentrations. Abt et al. (2000) found that cleaning (vacuuming, dusting, sweeping) and indoor work (walking around, field sampling, children playing) significantly increased concentrations of particles of 0.7 to 10 µm, in four Boston homes.

4.1.2 Resuspension from human body surfaces

Hession et al. (2006) investigated particle fall-off rates due to the degree of movement of a human with contaminated skin and also the effect of human body hair on particle fall-off. It was concluded that hairy skin retained particles for longer periods. Resuspension of particles from clothing, during human physical activity, has been examined in the medical, industrial and clean room industry. Cohen and Positano (1986) examined the work clothing of employees at a beryllium (Be) refinery to assess whether wear significantly affects the amount of Be-containing dust which resuspended. They found that old shirts resuspended significantly higher quantities of Be to the air than did new shirts and that a considerable fraction of the resuspended Be was respirable. Bohne and Cohen (1985) investigated resuspension from cotton and Nomex® aramid fabrics at a beryllium refinery and concluded that cotton resuspended a larger fraction of its contaminant load than Nomex®. In the medical industry, Loh et al. (2000) and Perry et al. (2001) found that staff uniforms were significant contributors to cross-contamination of formally deposited particles. Schneider (2008) found that clothing was a major contributor to ‘personal clouds’, which are particles generated by person-induced resuspension – i.e. particles shed from the person (skin scales, textile fibers, bacteria, etc), which are carried in the convective plume around the body. Clothing can also provide humans with a ‘shield’ from contamination. Andersson et al (2004) concluded

that when particles of 0.7 and 2.5 μm are deposited on clothing, less than 5% will penetrate through the clothing, thus reducing direct skin contamination by at least a factor of 20. A wide range of fabrics were tested, including thick and thin pure cotton, a thin 65% polyester and 35% cotton mix, and a thick 50% wool and 50% acrylic mix. An additional implication of the results of the study of Andersson et al (2004) is that if very few particles are penetrating through the clothing, then they have a potential for resuspension.

It is therefore well documented that human physical activity is a major contributor to particles resuspension from the human body surface (whether skin, hair or clothing) into the air surrounding the person engaging in the activity. If these resuspended particles are of a hazardous nature, they then pose a threat to a person's health. The mechanics by which surface residing particles can become resuspended and the specific forces involved will be discussed in section 5.

The enhancement of a person's inhalation exposure risk or the changing of the route of exposure from dermal to inhalation (via resuspension) is highly significant, as the dose associated with inhalation of hazardous aerosol particles is higher than is the case for dermal exposure. Andersson and Roed (2006) calculated estimates for the various contributions to dose that may have been received by people living in a dry-contaminated area in the Bryansk region, over 17 years after the Chernobyl accident. The estimated dose contribution from contamination of beta particles directly onto exposed skin was 0.2 – 0.3 mSv, while the dose contribution from inhalation of contamination was 6 – 8 mSv. Furthermore, contamination on the skin can be more easily removed by washing etc. than can contamination in the lungs, and the lung as a whole, has little regenerative capacity (Coggle et al., 1986) thus making it highly sensitive to damage by hazardous aerosols.

4.2 Contact Transfer

An additional transport process by which humans could become exposed to deposited hazardous particles is contact transfer. The process of surface-to-surface contact will typically involve human activity. For example, a person coming into contact with an aerosol particle contaminated surface, by, for example, sitting on a chair or resting their

arm on a desk, will transfer a portion of the deposited contamination to their clothing. Subsequent transfer of contamination is also possible, for example, if the person then comes into contact with a 'clean' surface; some fraction of the contamination on their clothing will transfer to the second surface.

Some published studies have demonstrated the spreading of contaminant via the process of contact transfer but aerosol mass transfer from one surface to another has been poorly quantified. Clothing has been identified as a significant source of contaminant transfer in the indoor environment. Tovey et al. (1995) state that dust mite allergen found in clothes could originate from many sources, including allergens collected when clothing items were laid on furnishings, floors and beds. De Lucca et al. (2000), Liccardi et al. (1998) and D'Amato et al. (1997) indicate that clothing is an important means of distributing cat allergen into cat-free environments. Cross-contamination by staff uniforms in the medical industry has also been examined. Loh et al. (2000) showed that the sleeves of medical students' white coats were more likely to contain bacterial colony counts than the backs of the coats. This is most likely due to contact transfer of particles from patients themselves, their clothing or bedding to the medical student as the patient is being examined. Mass transfer rates between skin and various other surfaces have also been documented. Reed (1975) studied the transmission of Rhinovirus colds through indirect contact and found that the virus could be spread from an infected person to another person via intermediate objects but that it was necessary to determine the efficiency of virus transfer from surface to surface.

Thus the process of contact transfer can result in dermal exposure to an initially uncontaminated person, and it can also increase a person's dermal exposure by transporting the contamination between indoor and human body surfaces. Furthermore, the redistribution of contamination via the process of contact transfer, can result in the contamination being available for further redistribution via other transport processes (e.g. contact transfer redistributing contamination from a table to skin, and subsequent resuspension from the skin), thus increasing a person's exposure risk by other exposure routes.

5 The Mechanics of Particle Resuspension

The mechanics of aerosol particle movement are notoriously difficult to precisely determine as they vary distinctly with particle characteristics (e.g. size), surface characteristics (e.g. roughness) and airflow characteristics (e.g. degree of turbulence) (Lai and Nazaroff, 2005). To aid in the analysis of the forces that contribute to aerosol particle resuspension, Figure 2 is a schematic diagram of the forces acting on a surface residing particle. The forces which aid resuspension are indicated in green, while those forces which hinder particle resuspension are shown in red.

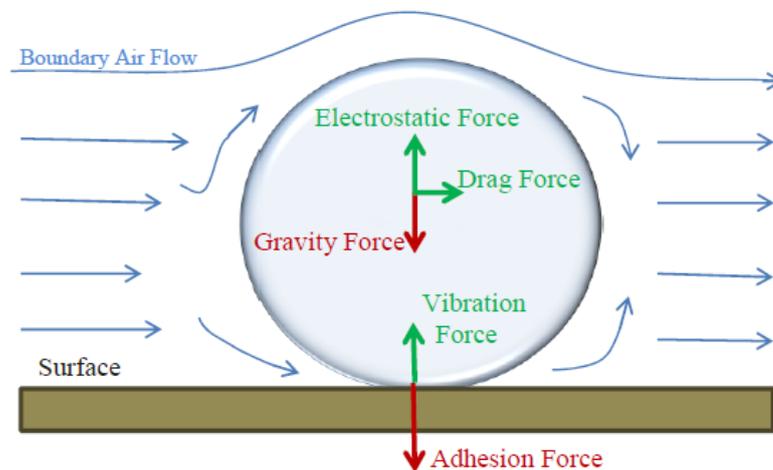


Figure 2. Schematic of the forces on surface residing particles

[adapted from Hu et al. (accessed Nov 2011)]

There are three main lift or resuspension forces associated with surface residing particles exposed to human activity (from Hu et al., accessed Nov 2011):

- *Mechanical Vibration Force*: This is the floor vibration response due to human activity, and is typically of magnitude 4 – 8 Hz.
- *Aerodynamic Force*: Air currents near a floor surface caused by human activity, can add drag forces to particles on the surface.
- *Electrostatic Force*: The amount of electrostatic charge depends on the material subjected to friction or separation, the amount of friction or separation, and the relative humidity of the ambient air. A strong electrostatic force can act on the charged surface particles to contribute to the resuspension force. The two main electrostatic forces are the Coulomb (field) and the image forces.

There are two primary forces, which attract a particle to a surface (attractive forces) thus preventing resuspension (from Hu et al., accessed Nov 2011):

- *Gravity*: Gravitation is a force via which all objects attract each other.
- *Adhesion Force*: The adhesion force exceeds the gravitational force and is therefore the dominant force that prevents particles from resuspending. The main adhesion forces include Van der Waals force (this is the largest), charge-induced electrostatic force and surface tension induced by an adsorbed liquid film. After initial contact, the adhesion force can gradually deform the contact parts, decrease the separation distance and increase the contact area until a new force equilibrium is reached.

If the lift forces are greater than the attractive forces for a particle on a surface, then the particle will be resuspended from that surface. The ease at which a contaminant can resuspend is primarily dependent on the characteristics of the resident surface material (Nicholson, 2009). However if the surface material remains constant, then the primary factor affecting resuspension is the particle size.

Many researchers have proven that large particles resuspend at a much higher rate than small particle, examples of which include (with resuspension specifically due to human physical activity indoors): Abt et al. (2000) and Ferro et al. (2004). This is because, for small particles, the ratio of its surface area in contact with the surface they are residing on, to its total surface area, is greater than the same ratio for large particles and therefore, it would take a greater force to resuspend smaller particles. This will be the case until particle size reaches a threshold diameter at which point the gravitational force will be the dominant force and particles will be restricted from resuspending fully (Hu et al., accessed November 2011). This threshold diameter below which particles can be fully resuspended into the atmosphere (at normal wind speeds) is at approximately 100 μm (Nicholson, 2009).

6 Objectives of the Present Work

The core of this thesis is structured as three journal papers, which individually address the gaps in resuspension and contact transfer knowledge, and within which the current state of knowledge is reviewed. The overall objectives of the current work are to examine the influence of five variables on the level of resuspension of hazardous aerosol particles from contaminated clothing. The variables to be investigated are listed below, the first four of which is described in detail in “*The Influence of Human Physical Activity and Contaminated Clothing Type on Particle Resuspension*”, and results of the investigation of the final variable is described in “*A Study of the Size Distribution of Aerosol Particles Resuspended from Clothing Surfaces*”. The variables include:

Physical Activity level: two levels of human physical activity are used - low and high- to determine and quantify any variation in the rate of particle resuspension.

Surface type: four different clothing material types are contaminated to determine if any specific material type hinders or enhances particle resuspension.

Body Location: the contaminated materials are attached to four different locations on the body and surveyed for variations in levels of particle resuspension.

Time: the rate, in real time, at which particles resuspend from the contaminated surfaces, is determined.

Particle Size: three different sized monodisperse particles are used to contaminate the surfaces and the size distribution of resuspended particles is examined.

Additionally, contact transfer rates between contaminated and clean surfaces are examined, and the findings are presented in detail in “*Mass Transport of Deposited Aerosol Particles by Surface-to-Surface Contact*”. Specifically, the objective is to quantify the mass transfer efficiencies of deposited aerosol particles to and from selected hard and soft surfaces typically found in a home or office. Other variables that are investigated include the applied pressure, contact time and contaminant loading. The goal of these experiments is to determine the range of transfer efficiencies that exist and to provide a preliminary investigation of the influence of several variables on these efficiencies.

7 Author Contribution to Published Papers

- **McDonagh A.** and Byrne. M. A. 2013. *The Influence of Human Physical Activity and Contaminated Clothing Type on Particle Resuspension*. Journal of Environmental Radioactivity, (in press). DOI: 10.1016/j.jenvrad.2013.10.012.

Ann McDonagh developed the experimental procedures, conducted the experiments, collected the data, analysed the data, and wrote the paper. She received support from her PhD supervisor Dr. Miriam Byrne.

- **McDonagh. A.** and Byrne M. A. *A Study of the Size Distribution of Aerosol Particles Resuspended from Clothing Surfaces*. (submitted for publication in Indoor Air).

Ann McDonagh developed the experimental procedures, conducted the experiments, collected the data, analysed the data, and wrote the paper. She received support from her PhD supervisor Dr. Miriam Byrne.

- **McDonagh. A.,** Sextro. R. G., Byrne. M. A. 2012. *Mass Transport of Deposited Aerosol Particles by Surface-to-Surface Contact*. Journal of Hazardous Materials. Volumes 227-228, pp. 370-377. DOI: 10.1016/j.jhazmat.2012.05.070.

Ann McDonagh developed the experimental procedures, conducted the experiments, collected the data, analysed the data, and wrote the paper. She received support from her PhD supervisor Dr. Miriam Byrne and also from Dr. Richard Sextro at the Lawrence Berkeley National Laboratory, where the experimental work was carried out.

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The Influence of Human Physical Activity and Contaminated Clothing Type on Particle Resuspension

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Abstract

A study was conducted to experimentally quantify the influence of three variables on the level of resuspension of hazardous aerosol particles from clothing. Variables investigated include physical activity level (two levels, low and high), surface type (four different clothing material types), and time i.e. the rate at which particles resuspend. A mixture of three monodisperse tracer-labelled powders, with median diameters of 3, 5, and 10 microns, was used to “contaminate” the samples, and the resuspended particles were analysed in real-time using an Aerodynamic Particle Sizer (APS), and also by Neutron Activation Analysis (NAA).

The overall finding was that physical activity resulted in up to 67 % of the contamination deposited on clothing being resuspended back into the air. A detailed examination of the influence of physical activity level on resuspension, from NAA, revealed that the average resuspended fraction (RF) of particles at low physical activity was 28 ± 8 %, and at high physical activity was 30 ± 7 %, while the APS data revealed a tenfold increase in the cumulative mass of airborne particles during high physical activity in comparison to that during low physical activity. The results also suggest that it is not the contaminated clothing's fibre type which influences particle resuspension,

but the material's weave pattern (and hence the material's surface texture). Investigation of the time variation in resuspended particle concentrations indicated that the data were separable into two distinct regimes: the first (occurring within the first 1.5 minutes) having a high, positive rate of change of airborne particle concentration relative to the second regime. The second regime revealed a slower rate of change of particle concentration and remained relatively unchanged for the remainder of each resuspension event.

1 Introduction

Following an accidental or deliberate release of hazardous materials, such as radioactive (e.g. Fukushima) or infectious species (e.g. H1N1 virus) to the atmosphere, there is a risk of exposure of large population groups and individual persons who reside both indoors and outdoors. In preparing for this scenario, accurate estimates of whole body exposure arising from all exposure pathways are necessary in order to design effective countermeasures. Current dosimetric models, for example, that of Andersson et al. (2004), focus on aerosol inhaled while initially airborne, with some reference to particles deposited on the human body, but secondary exposure and inhalation is not considered. One secondary exposure pathway that merits investigation, and for which no comprehensive experimental data are available, is re-exposure to aerosol contaminant that was formerly deposited on clothing and other indoor surfaces. This is especially important in the case where a person might be unwittingly contaminated and might spread this contamination to others via the process of resuspension. These exposure pathways may also have significance for airborne pollutants that are non-radioactive, e.g. infectious aerosol that becomes re-entrained from disturbance of hospital bedding, etc.

Numerically, resuspension rates are low relative to deposition rates (e.g. Thatcher and Layton, 1995) but the concentration of resuspended contaminated particles can significantly influence a persons' exposure. The size distribution of resuspended particles is different from the size distribution of depositing particles, with larger particles being more readily resuspended (Andersson et al., 2004, Ferro et al., 2004).

A wide range of *contaminated surfaces* have been investigated in the context of particle resuspension due to human physical activity, but resuspension studies focus on quantifying the level of resuspension from flooring surfaces (Ferro et al, 2004, Long et al, 2000, Abt et al, 2000); contamination on, and its removal from, clothing surfaces has received little attention, apart from a few studies in the medical devices and cleanroom industry (Cohen and Positao, 1986; Bohne and Cohen, 1985). The removal of particles from human skin has been investigated; Hession et al (2006) studied the effect of human body hair on particle fall-off and concluded that hairy skin retained particles for longer periods. To supplement the existing knowledge base, the aim of the present study is to quantify the level of resuspension of hazardous particles directly from clothing surfaces

worn by a physically active human, who is engaging in various levels of typical daily physical activity. Earlier studies have generally determined particle resuspension rates and factors based on changes in the total airborne particulate concentrations, which is not chemically specific and so could not directly isolate the resuspended particles from others that were airborne. While the current study also uses this accepted experimental method, an additional and more comprehensive method, using Neutron Activation Analysis (NAA), will be used to calculate the fraction of originally deposited particles which resuspended, directly from the clothing surface.

2 Materials and Methods

A summary of the experimental strategy for studying particle resuspension from clothing surfaces is as follows: the material surfaces to be ‘contaminated’ were first exposed to aerosol particles in the form of tracer labelled silica powder. They were subsequently attached to a volunteer who engaged in a predefined physical activity for a specified length of time. Samples of the material were analysed both before and after the resuspension process to determine the proportion of the original deposited mass which became resuspended. The air in the resuspension chamber was continuously monitored to determine the size spectrum of resuspended particles in the air.

2.1 Aerosol Particles

The particles used throughout these experiments were monodisperse silica particles (Alltech) labelled with a rare earth metal tracer. Three particle sizes, with manufacturer specified mean diameters of 3 μm , 5 μm and 10 μm , were chosen for their similarity to that of the refractory group of radioactive aerosols released into the atmosphere following a nuclear explosion and also to that of some biological species e.g. anthrax. It is important to note that although the particles were monodisperse, each batch of particles will have a size distribution about their mean diameter. The three sizes were labelled with Europium (EuCl_3), Dysprosium (DyCl_3) and Indium (InCl_3) respectively following the method described by Jayasekera et al. (1989). For Dysprosium, the average labelling yield was 5.20 mg of Dy on 1 g of labelled silica. To reduce the

number of experimental replications that needed to be carried out and also the cost of analysis, the three particle sizes were mixed together for deposition onto the surfaces for all experiments.

2.2 Aerosol Deposition

All clothing samples to be used in the resuspension experiments were first contaminated with the silica particles in a 2.25 m³ aluminium deposition chamber. Inside the chamber a small 2W fan was mounted 0.2 m from the chamber ceiling (chamber height = 1.5 m), centrally aligned and orientated vertically downwards, to simulate real room mixing conditions. An Aerodynamic Particle Sizer (APS) was operational during the deposition events.

The particles were injected into the chamber using a dry powder particle generator (Palas RBG-1000) located on the roof of the chamber. In all cases, the aerosol was passed through a tube containing an array of 12 x 33 KBq Am241 radioactive sources; using the calculations of Cooper and Reist (1973) it was estimated that, for the aerosol flow rate used, the ratio of the residence time of the aerosol in the tube and the characteristic source strength used was greater than unity, so that aerosol charge equilibrium would be reached.

Before commencing the 'contamination' of fabric samples, the floor of the deposition chamber was analysed for any variations in the spatial uniformity of particle deposition; a variation of only 9 % was observed over a central floor area of 43 x 76 cm.

2.3 Clothing Surfaces

As the human body is typically covered with some degree of clothing, four common clothing materials were chosen for the experiments: cotton, polyester, fleece and denim. The chosen materials consisted of natural and synthetic fibres with smooth and rough surface textures. The natural fibre used was a 100 % cotton material. It had the same tight weave pattern as the 100 % polyester material which is a synthetic fibre, as observed by electron microscopy. Fleece is also made from 100 % polyester but has a

much rougher surface texture resulting from the disordered nature of the weave. A denim material was chosen due to its popularity as a clothing textile and the particular denim used in this work consisted of 65 % cotton, 32 % polyester and 3 % spandex. As these materials were composed of different fibres and colours, they were pre-tested by Neutron Activation Analysis (NAA) to ensure they did not contain elements that would interfere with the analysis of the tracer particles to be deposited onto them.



Figure 1. Specific locations of contaminated samples on volunteer

Large samples of each material type were contaminated for each experimental run. This was to ensure there would be a sufficient level of contaminant available on the volunteer during the resuspension process, to be resuspended and thus captured by the aerosol sampling equipment. Contaminated material samples were placed onto the volunteers' legs, arms and chest, as can be seen in figure 1. The leg pieces were 25 x 40 cm and were wrapped around each lower leg, thus covering almost all of the lower legs (front and back) between the knee and ankle. The chest piece was 20 x 20 cm and the arm pieces were 15 x 30 cm, one each on the outer arm between the shoulder and elbow. The clothing samples had safety pins attached to their corners before contamination. This was to facilitate quick and easy attachment of the contaminated samples to the volunteer (by an assistant), with a minimum of disturbance to the samples themselves. Each of the large clothing samples also had two smaller pieces of the same material (ranging from 1 x 4

cm to 2 x 5 cm, depending on the material type) pinned onto them before contamination. These were both analysed by NAA, one before and one after the resuspension process, for reasons which will be discussed in section 2.5. The size of the NAA capsules restricted the size of material sample which could be analysed.

2.4 Aerosol Resuspension

After contamination, the clothing samples were placed into individual, sealed and labelled plastic containers and transported to the vicinity of the resuspension chamber, a purpose-built 15 m³ plywood test room. Inside the chamber were two APS's, one at ankle height (15 cm from the floor) and the other at waist height (90 cm from the floor). In the centre of the chamber floor was a grounded metal plate upon which the volunteer stood while performing the experiment. This was to ensure that any static charge generated by the movement of the volunteers' feet, would not influence the behaviour of the resuspended particles. Also, the smooth surface texture of the metal made it easier to ensure complete decontamination between experiments.

Directly outside the chamber, the volunteer dressed in a clean room suit (Macrobond hooded coverall from Caulfield Industrial), booties (Polylatex shoecover 4 g from Caulfield Industrial), gloves (powder free purple nitrile) and a mask (8835 FFP3D disposable mask from Anderco Safety). The cleanroom suit was chosen for its non-linting and anti-static properties, so that it can be assumed that any airborne particles detected by the APS's were as a result of particles resuspending from the contaminated material and not from the volunteer's personal clothing. An assistant then carefully removed one of the four types of 'contaminated' clothing samples from their boxes and pinned them to the volunteer's clean room suit. The placing of clothing samples over the clean room suit will generate an effect similar to that of a person wearing multiple layers of clothing (denim jeans over underwear, jumper over t-shirt etc.).

When fully attired and with samples attached, the volunteer entered the resuspension chamber and participated in one of two pre-defined physical activity patterns: low physical activity – walking at a pre-defined rate for 20 minutes, or high physical activity – Irish dancing to a Reel (Wikipedia, accessed Nov 2011) with added swinging of the arms for 10 minutes (chosen to facilitate the accurate reproduction of movement in each

repeated experiment). The duration of the resuspension event for high physical activity was half that for low physical activity as the level of physical exertion required was not possible to keep constant over a longer period of time. The volunteer then exited the chamber and the assistant removed the clothing samples and placed them back into their individual boxes. The resuspension chamber and all equipment were thoroughly cleaned between experiments by washing with water and using alcohol wipes. Additionally, a multi-functional air purifier (model XJ-3000C by Heaven Fresh) with a HEPA filter, carbon filter, ioniser and sanitiser was operational between experiments, to ensure the air was completely devoid of any tracer particles.

2.5 Analysis

The primary aerosol measuring instrument used in this study was the Aerodynamic Particle Sizer (APS) [Model 3321 by Trust Science Innovation (TSI)]. The APS sizes airborne particles in the range of 0.5 to 20 μm in aerodynamic diameter. Throughout these experiments, the concentration and size distribution of the airborne particles in both the deposition and resuspension chambers were continuously measured in real time using the APS's. The second analysis technique employed was Neutron Activation Analysis (NAA). The surfaces contaminated with labelled silica were analysed by NAA at the Reactor Institute in the Delft University of Technology, the Netherlands. NAA allows discrete sampling of elements as it disregards the chemical form of a sample, and focuses solely on its nucleus. The three rare earth metal labels used in these experiments have a half-life of 9.3 h, 2.3 h and 54 min for Eu, In and Dy respectively and they decay as follows: $^{151}\text{Eu} (+ \text{neutron}) = ^{152\text{m}}\text{Eu} (- \gamma) = ^{152}\text{Gd}$, $^{164}\text{Dy} (+ \text{neutron}) = ^{165}\text{Dy} (- \gamma) = ^{165}\text{Ho}$, $^{115}\text{In} (+ \text{neutron}) = ^{116\text{m}}\text{In} (- \gamma) = ^{116}\text{Sn}$. The third analysis technique used was Scanning Electron Microscopy (SEM).

3 Results and Discussion

3.1 Airborne Mass Concentration Variation with Time, During Resuspension

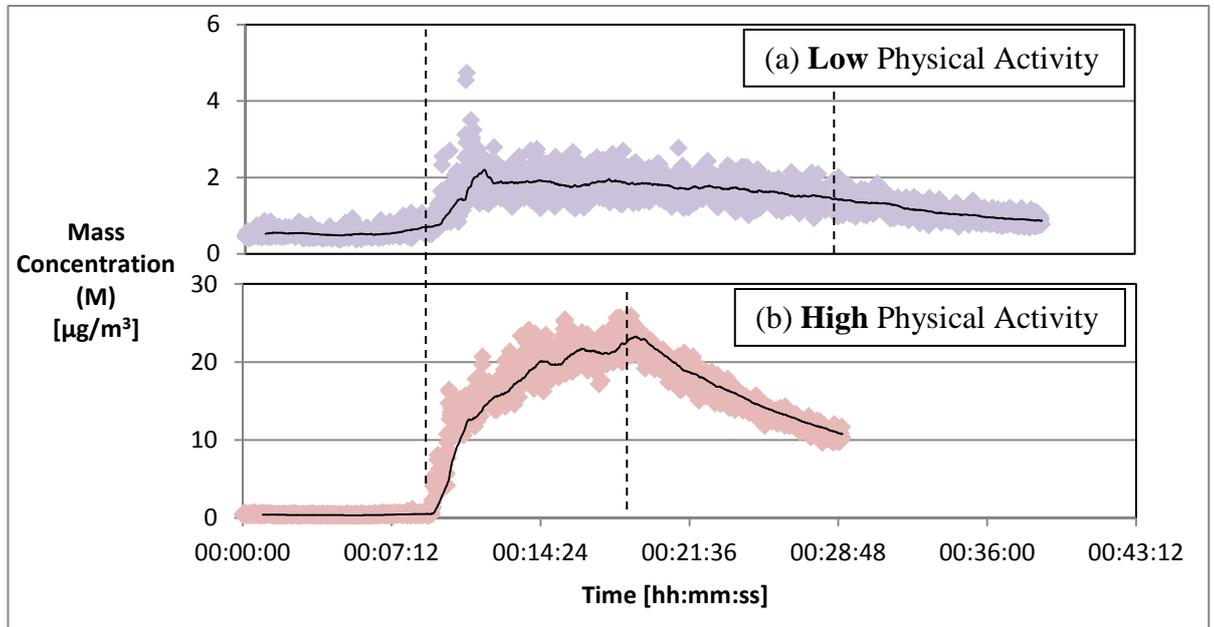


Figure 2. Mass concentration of airborne particles during resuspension due to (a) low physical activity and (b) high physical activity, as a function of time

Figure 2 indicates the change in particle mass concentration with time (as detected by an APS in the resuspension chamber) before, during and after a person wearing contaminated clothing is engaging in (a) low physical activity and (b) high physical activity. Each data point represents the cumulative sum of the airborne mass concentration over all particle sizes, at a particular second and the trendline indicates the one minute average. The vertical dashed lines indicate the instances when the contaminated person entered and exited the resuspension chamber i.e. the start and end of the resuspension event. During low physical activity, the contaminated person was physically active within the resuspension chamber for 20 minutes and for 10 minutes during high physical activity.

It is clear from figure 2(a), that there is an immediate and steady increase in airborne particle concentration from the moment the person enters the chamber. The averaged cumulative concentration reaches a peak of $2.26 \mu\text{g m}^{-3}$ just 1.5 minutes after the resuspension event begins. The particle concentration then rapidly decreases to $1.81 \mu\text{g}$

m^{-3} in a further 30 seconds. After that point (2 minutes into the resuspension event) the airborne particle concentration slowly decreases. A linear trendline (best fit to data) plotted to the airborne particle concentration readings for the last 18 minutes of the resuspension event indicates a decreasing slope: i.e. a decrease in the rate of change in airborne particle concentration of $-0.026 \pm 0.002 \mu\text{g m}^{-3} \text{min}^{-1}$.

The pattern shown in figure 2(b) is similar to figure 2(a): there is an immediate and steady increase in airborne particle concentration from the moment the person enters the chamber. However, the difference in pattern for figure 2(b) is that the airborne particle concentration now continually increases throughout the entire 10 minutes of the resuspension event. The peak averaged concentration of approximately $23.2 \mu\text{g m}^{-3}$ occurs at the end of the resuspension event, just as the contaminated volunteer exits the chamber.

The rate of change of airborne particle concentration during resuspension for both physical activity levels can be split into two separate regimes, the first occurring during the initial 1.5 minutes, and the second over the remainder of the resuspension event. For the first regime - during the first 1.5 minutes of the resuspension event - both physical activity levels result in an increase in airborne particle concentration but high physical activity results in a rate of increase four times that corresponding to low physical activity, i.e. $4.5 \pm 0.5 \mu\text{g m}^{-3} \text{min}^{-1}$ and $1.1 \pm 0.2 \mu\text{g m}^{-3} \text{min}^{-1}$, respectively.

For the second regime – during the remaining period of each resuspension event – the change in airborne particle concentration shows an opposing trend for low compared to high physical activity levels. During low physical activity, the particle concentration decreases at a comparatively slow rate of $-0.026 \pm 0.002 \mu\text{g m}^{-3} \text{min}^{-1}$, while it increases and at a more rapid rate of $1.00 \pm 0.03 \mu\text{g m}^{-3} \text{min}^{-1}$ during high physical activity.

Wu et al. (1992) noted a very similar pattern in resuspension with time when examining the resuspension of uranine particles, polymer microspheres, lycopodium spores and Johnson grass pollen from glass, plexiglass and white oak leaves at windspeeds of 4 to 8ms^{-1} . They concluded that the resuspension rates varied over time, defined by two separate regimes. The first regime occurred in the first minute of resuspension and was characterised by higher resuspension rates (0.17s^{-1}). After the first minute, particles

which are easily resuspended had been removed, leaving only particles with much smaller resuspension rates (0.029 s^{-1}) for the second regime. The noting of two resuspension regimes is identical to this current study and also the time frame for the first regime is similar, with Wu et al. (1992) noting 1 minute and this study observing 1.5 minutes. However the slower resuspension rate during the second regime as determined by Wu et al. (1992) is comparable only to the decrease in the rate of change of airborne particle concentration detected in this study at low physical activity. High physical activity resulted in an increase in the rate of change of airborne concentration during the duration of the second regime (albeit at a slower rate in comparison to the first regime).

Hession et al. (2006) compared the clearance of fluorescent 3 and 10 μm particles from skin during two different activity levels: low activity where the volunteer remained seated indoors, and high activity where the volunteer engaged in vigorous outdoor walking in dry, breezy conditions. The rates of clearance of particles from the skin with time during low activity was shown to follow an exponential pattern, however a very different clearance pattern was displayed during high activity. Based on figure 3 in Hession et al. (2006), during high activity, the mass of particles remaining on the volunteer's skin reduced by approximately 88 % in the first 30 minutes and then remained approximately constant for the remaining 170 minutes. This again displays a two regime removal of particles from a surface with time and also exhibits a difference in the particle clearance pattern between two different physical activity levels. This will be elaborated upon in section 3.2.

3.2 Resuspension with Varied Physical Activity Level

The purpose of this section is to examine the resuspended fraction (RF) data for each particle size, at low and high physical activity levels. The RF is calculated by measuring the difference in mass concentration of particles deposited on the material sample before and remaining on the material sample after the period of resuspension, as a percentage of the mass concentration deposited on the material sample before resuspension. The data is averaged over all contaminated material types. Low physical activity consisted of walking for 20 minutes. High physical activity was representative of running and simulated by modified Irish dancing to a Reel for 10 minutes, as described in section 2.4.

The NAA data indicates that the fraction of particles which are resuspended from the clothing of a person engaged in low physical activity ranges from 8 to 52 %, with an average RF across all sized particles of 28 ± 8 %. During high physical activity, between 3 and 67 % of particles formerly deposited on various clothing types was found to resuspend, with an average RF value across all particle sizes of 30 ± 7 %. A two-tailed paired T-test shows that the difference between the average resuspended fraction (averaged over all particle sizes and material types) at low and high physical activity level is not statistically significant.

Figure 3 shows the average size distribution of particles in the air surrounding a contaminated person engaging in either low (blue diamonds) or high (red circles) physical activity, as measured by an APS at waist height. The mass distribution is calculated by averaging the total concentration per second in each size range, over the entire physical activity (i.e. resuspension) duration: 20 minutes for low and 10 minutes for high physical activity. This is calculated for each repeated experiment and for each material type (i.e. 16 individual experiments) and an overall average is obtained for each physical activity level.

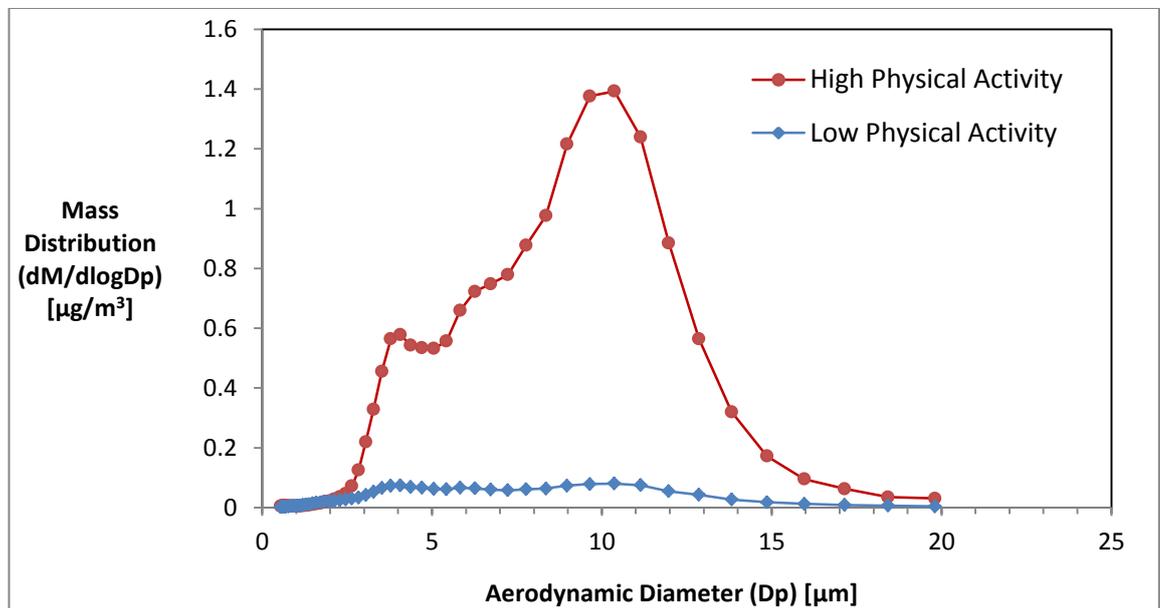


Figure 3. Size distributions of particles during low and high physical activity

The sum of the mass concentration of airborne particles over all particle sizes during low physical activity is $1.72 \mu\text{g m}^{-3}$ and during high physical activity is $16.93 \mu\text{g m}^{-3}$. This is almost a tenfold increase in the cumulative mass of airborne particles during high

physical activity in comparison to that during low physical activity. This difference is statistically significant, with a p-value of 0.000001 based on a two tailed, paired student t-test. Therefore, within the range of particle sizes measured, and between these two physical activity levels, there is a significant difference in the mass of airborne particles resuspended from a contaminated persons' clothing. However, this conclusion is drawn from the APS data and appears to contradict the results from the NAA data. This apparent contradiction is due to the capturing of different features of the resuspension process by the two different analytical techniques, as discussed below.

The APS data shows the airborne mass concentration as a function of particle size and thus measures all particles that are airborne, regardless of their origin. NAA data are used to determine the resuspended fraction (RF), which is calculated by measuring the difference in mass concentration of particles deposited on the material sample before and remaining on the material sample after the period of resuspension, as a percentage of the mass concentration deposited on the material sample before resuspension. Hence the concentration of particles remaining on a sample after resuspension is the key variable in determining the resuspended fraction from that sample. However, this value may be higher than expected due to an unquantified number of particles remaining on that sample after the resuspension event, caused mainly by three processes which are described below.

The first contributor to a higher than expected mass concentration of particles remaining on the material sample after the resuspension event is impaction of resuspended particles back onto the material sample. During resuspension, the volunteer wearing the contaminated clothing is moving vigorously on a fixed position on the floor for a period of time. Thus some particles which are initially resuspended may be re-deposited onto the clothing sample due to impaction, as the body part is moved through the 'cloud' of resuspended particles. The movement of a body part through a particle-laden airstream which results in impaction, is analogous to the process of particle impaction when a moving particle-laden airstream encounters a stationary object – this process has been well described (e.g. Golovin and Putnam, 1962, Hinds, 1999, Lange, 1995, and Ranz and Wong, 1952).

The second contributor is enhanced deposition due to both increased air velocities around the moving person and the rate of displacements of the body parts (and hence the

material samples) themselves. Wu et al. (1992) concluded that, once particles are resuspended from their residing surface, their trajectories depend on the characteristics of the turbulent air flow and not on the initial velocity of the particle. The resuspended particles in this study will have a high velocity due to the increased air velocity caused by the physical movement of the contaminated volunteer. Rim and Novoselac (2009) examined the influence of localised arm movement on air velocities using a mechanical manikin sitting on a chair and the hands were subjected to large and small motion. The large motion was a periodic rotation of the entire arms back and forth by $\pm 30^\circ$ (represents active hand movement e.g. filing), the small motion was a periodic up and down rotation of the elbow by $\pm 10^\circ$ (represents limited hand movements e.g. typing). They concluded that physical activity of the manikin does significantly affect the air velocity at 20 cm above the floor but does not largely affect the air velocity above the head and also that large motion has a greater affect than does small motion. Bjorn and Nielsen (2002) also investigated the influence of the physical activity of a manikin in a room and found that a larger degree of air mixing occurred in the room with the moving manikin than with a stationary one. The volunteer in the current experiments is moving at a fast pace, for example, during high physical activity (modified Irish dancing) the volunteer jumps from one leg to the other at an average rate of 55 jumps per minute which corresponds to a rate of leg displacement of approximately 0.75 m s^{-1} and a rate of arm displacement of approximately 0.5 m s^{-1} . This movement will undoubtedly increase the air movement around the active contaminated person and thus increase the resuspended particles' velocity which will lead to enhanced deposition back onto the contaminated material as the material is repeatedly moved through the 'cloud' of resuspended particles.

The third contributor is the penetration of resuspended particles deep into the weave of the material. Because of the particles' increased velocities when they redeposit, they will have an increased kinetic energy and hence may penetrate deeper into the weave pattern of the material. These particles will therefore be more difficult to re-resuspend. This point will be elaborated in section 3.3.

As a consequence of the above three events occurring during resuspension, the mass of particles calculated to be on the clothing sample after the period of resuspension may be higher than would be the case if all particles which initially resuspended were removed

and not available to re-deposit. Therefore a lower RF than is actually the case is calculated but it should be noted that this RF is representative of a real-life scenario where impaction and secondary resuspension would be continuously occurring.

The rate of recapture is difficult to quantify, as it is dependent on a number of variables including: air velocity, target velocity, particle size and Stokes number, the boundary layer thickness and Reynolds number. The impact efficiency onto a cylinder for particles with very small Stokes numbers (as is the case here) is difficult to assess as these particles are extremely sensitive to the boundary layer very close to the cylinder (Haugen and Kragset, 2010). According to figure 3 of Wessel and Righi (1988), for a Reynolds number of zero (in this experiment, $Re \sim 0.2$ around the arm) and a Stokes number of 1.5, the target efficiency will be 50 %. The APS data indicates a 10 fold increase in the *mass* of airborne particles but only a two fold increase in the *number* of airborne particles, during low and high physical activity. Hence, a greater proportion of the smaller particles are being recaptured by the clothing, particularly at higher physical activity levels. This is due to the smaller particles being more likely to remain in the air streams created by the moving body part and thus being intercepted by the body part moving back through the same air location. The bigger particles will leave the air streams and move away from the body part, and are more likely to be captured by the APS or deposit to the floor etc. This is confirmed by the NAA data, where the RF increases with increasing particle size at both physical activity levels but to a greater extent at high physical activity (this will be discussed in greater detail in a succeeding journal article).

3.3 Resuspension with Varied Contaminated Clothing Material Type

Four different material types were investigated – cotton, polyester, fleece and denim. Each material type was contaminated and worn by a volunteer engaging in physical activity. The concentration of particles resuspended from each material type was investigated, with each material type experiment repeated four times and an average distribution calculated. This was possible as the consistency between data derived from repeated experiments using any one material type was high (all p values ≥ 0.17 , i.e. no significant difference).

Figure 4 shows the average mass distribution of airborne particles as a function of particle size, for each material type. The average mass distribution is averaged over the entire resuspension process (10 mins at high physical activity) and over all four repeated experiments of each material type. The size distribution curve for particles resuspended from fleece is represented with blue diamonds, polyester with red squares, denim with green triangles and cotton with yellow circles.

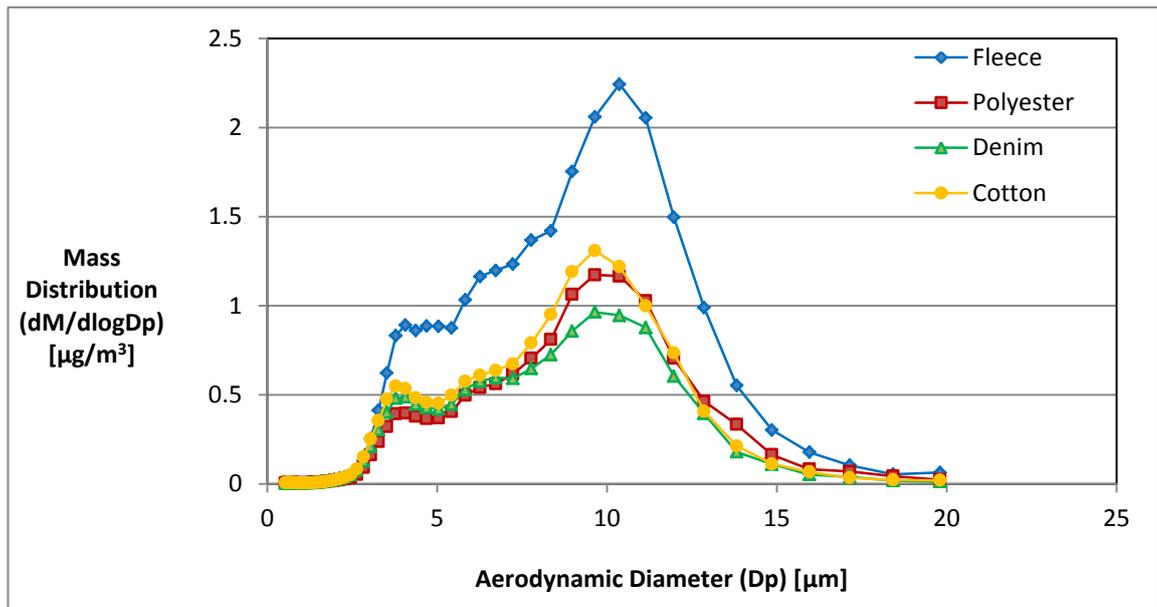


Figure 4. Size distribution curve of particles resuspended from all four material types, during high physical activity

From figure 4 it can be seen that the size distribution curve for particles resuspended from fleece corresponds to a higher mass concentration than that for the three other material types. The cumulative mass concentration of particles of all sizes resuspended from fleece is $26.28 \mu\text{g m}^{-3}$, from cotton is $15.13 \mu\text{g m}^{-3}$, from polyester is $13.47 \mu\text{g m}^{-3}$ and from denim is $12.84 \mu\text{g m}^{-3}$. Analysing the distributions using a student t-test (two tailed, two samples of equal variance) identifies fleece as being significantly different from the distributions of all other material types, with $p = 0.02$, 0.01 and 0.04 between fleece and polyester, denim and cotton, respectively. However, the distributions of polyester, denim and cotton are not significantly different from each other.

The above finding is confirmed using the data analysed by NAA for the same set of data (figure 4). The average RF for fleece is $41 \pm 10 \%$, for polyester is $31 \pm 7 \%$, for denim is $28 \pm 4 \%$ and for cotton is $25 \pm 5 \%$. Using the same statistical test as was used for the

APS data, the resuspended fraction of original contaminant from fleece and fleece alone is significantly different from that of each of the three other contaminated material types, with $p = 0.03$, 0.005 and 0.001 between fleece and polyester, denim and cotton, respectively.

The above finding may be explained by considering the varied surface texture of the material types. The synthetic fleece used in these experiments is made from 100 % polyester as is the polyester material. However the weave pattern is very different for both material types, as confirmed using SEM imagery (figure 5). The polyester material has a very uniform and tight weave pattern (which, as an aside, is found to be very similar to the weave pattern for membrane collection filters, described by Bénesse et al., 2006) while the fleece is more irregular and loose.

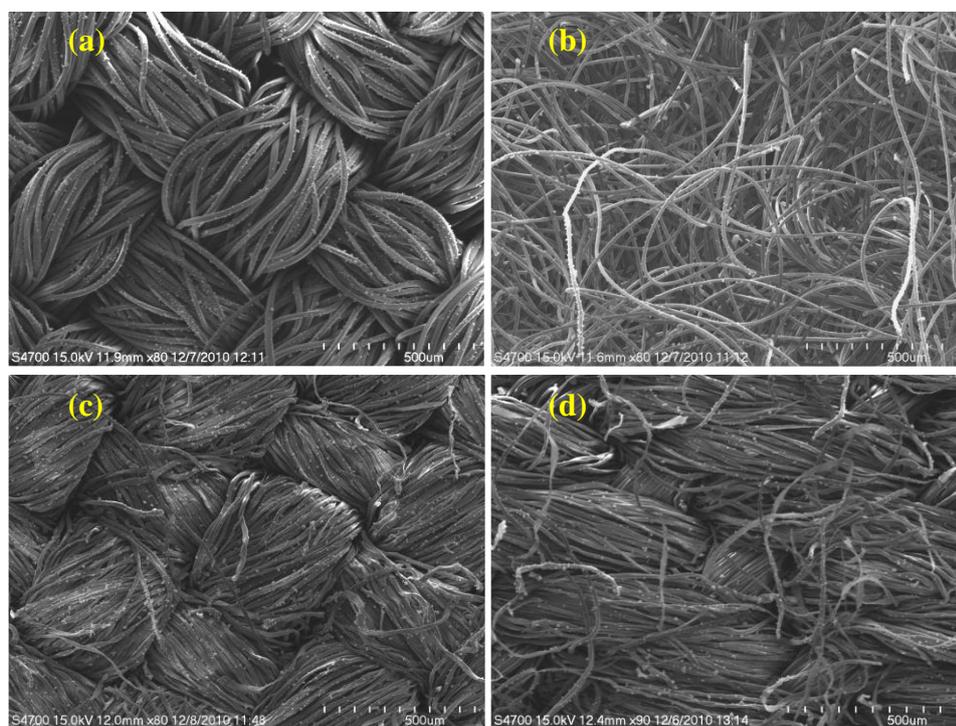


Figure 5. SEM images showing the weave pattern of: (a) polyester (b) fleece (c) cotton and (d) denim. Each image is taken at 80 or 90 times magnification and the scale at the bottom right corner is 500 μm .

Although the majority of deposited particles reside in a monolayer on the surface layer of the material (as verified using SEM imagery of contaminated surfaces), some particles deposited onto the polyester surface penetrate into the crevices between weave strands and could potentially be trapped there (due to the tightness of the weave) and thus be harder to resuspend. An example of particles deposited deep into the weave

can be seen in figure 6. This could account for the degree of resuspension of particles from polyester, cotton and denim (all with the same weave pattern) to be lower than that from fleece.

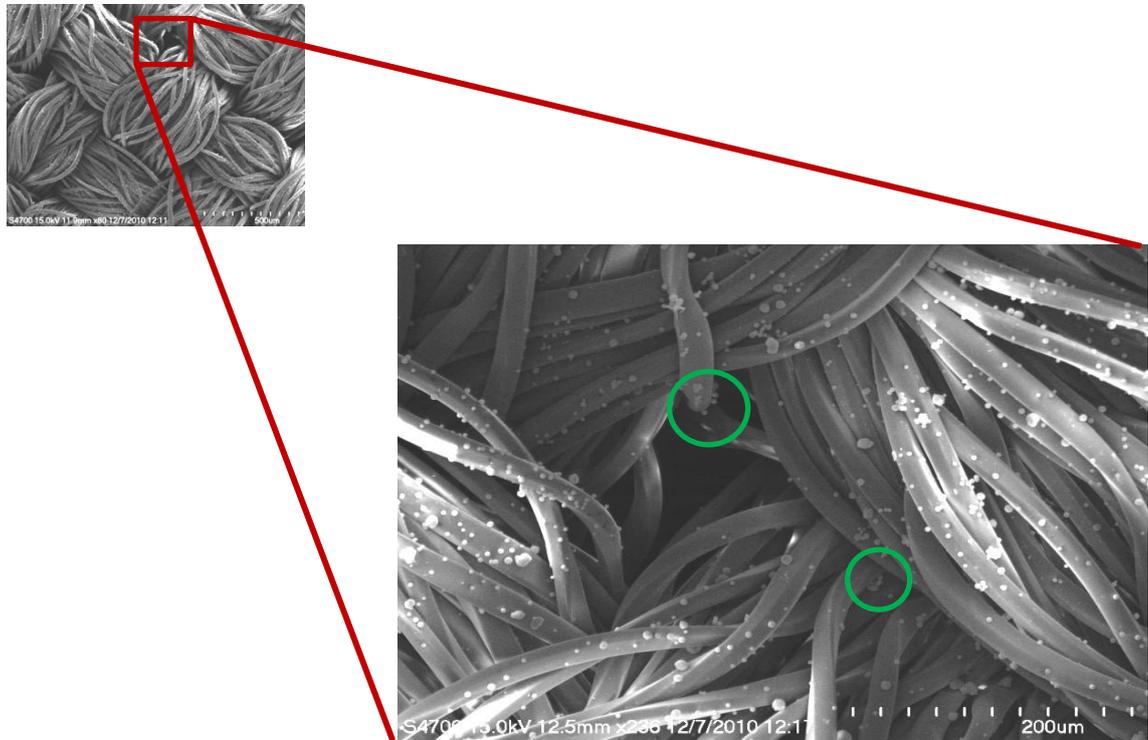


Figure 6. SEM image of particles deposited deep into the weave (circled areas) of contaminated polyester

The APS results in figure 4 and the succeeding discussion of the related NAA data, also indicate that the fibre used to make the cloth or material does not significantly affect resuspension, as the polyester and cotton materials used in these experiments were found statistically to have the same level of resuspension from them. While they are made from different fibres - polyester is synthetic and cotton is natural - they both have the same weave pattern. This indicates that it is the surface texture roughness and not material type, which is the more important factor in relation to the resuspension of hazardous aerosol particles from clothing surfaces.

Previous findings indicate that increasing surface texture roughness results in lower amounts of particle resuspension, for example, Wu et al. (1992), Braun et al. (2002), Hession et al. (2006) and Andersson et al. (2004), and this appears to contradict the findings in the present work. However, the apparent discrepancy can be explained by

considering the relative significance of macroscopic and microscopic features of the fabric. Macroscopically, fleece would have a rougher surface texture than polyester and should therefore demonstrate a lower resuspended fraction which it does not. However, microscopically, the polyester material has a rougher surface texture than the fleece material, due to their respective weave patterns. If one considers that penetration and settling of particles deep into the weave of the clothing fabric (which is occurring in the current experiments due to the increased air velocities around the volunteer from their physical movement and also due to impaction of airborne particles back onto the clothing samples), then the significance of the microscopic features of the fabric can be realised.

One parameter which may have an influence on particle behavior but which was not investigated in these experiments is static charge. The electrostatic forces associated with surface residing particles, can both aid and hinder particle resuspension and are dependent on a number of factors including the composition of the material subjected to friction, the amount of friction or surface agitation, and the relative humidity of the ambient air.

4 Conclusions

The influence of human physical activity level on particle resuspension from clothing was examined using two analysis techniques: Neutron Activation Analysis (NAA) and Aerodynamic Particle Sizing (APS). The NAA data showed that the average resuspended fraction (RF) of particles at low physical activity was $28 \pm 8 \%$, and at high physical activity was $30 \pm 7 \%$, a statistically insignificant difference. In contrast, the APS data revealed a statistically significant tenfold increase in the cumulative mass of airborne particles during high physical activity in comparison with that during low physical activity. While both techniques indicate an increase in resuspension with increasing physical activity level, the difference in terms of statistical significance is attributed to the two analysis techniques capturing different aspects of the resuspension process, as discussed in section 3.2.

In respect to the influence of the contaminated surface type on particle resuspension, the results obtained indicate that it is not the contaminated clothing's fibre type which

influences particle resuspension, but the material's weave pattern (and hence the material's surface texture). Fleece was found to exhibit the highest levels of particle resuspension (cotton, denim and polyester showed no significant difference), as particles did not become trapped deep within the weave pattern of the material and were thus relatively easily released from the non-uniform weave, when the material was subjected to agitation.

An investigation of the change in airborne mass concentrations with time, due to the resuspension of particles from contaminated clothing, revealed that the data was separable into two distinct regimes: the first having a high, positive rate of change of airborne particle concentration relative to the second regime, and occurring within the first 1.5 minutes of the beginning of the resuspension event. The second regime revealed a slower rate of change of particle concentration and remained relatively unchanged for the remainder of each resuspension event.

In conclusion, based on the variables investigated in this study, the conditions for which the highest fraction of initially deposited particles became resuspended were during the wearing of contaminated clothing of fleece material by a person engaged in a high level of physical activity, and during the first 1.5 minutes of that activity. The finding that physical activity can cause up to 67 % of contamination formerly deposited on clothing to be resuspended back into the air implies that the transport process of resuspension is of considerable importance for accurate determination of human exposure risks.

5 Acknowledgements

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*A Study of the Size Distribution of
Aerosol Particles Resuspended from
Clothing Surfaces*

(Research paper submitted for publication in Indoor Air)

A Study of the Size Distribution of Aerosol Particles Resuspended from Clothing Surfaces

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Abstract

A primary factor governing the impact of hazardous aerosol particles on the human body is the size of the contaminant particles. Secondary exposure to humans can occur when initially deposited particles are re-entrained into the air via the transport process of resuspension. This study has experimentally investigated the size distributions of a mixture of monodisperse 3, 5, and 10 micrometre particles after they were collectively deposited on, and then resuspended from the clothing of a contaminated person engaged in varying degrees of physical activity. The generally accepted theory, that the likelihood of particles resuspending from a surface will increase with increasing particle size, has been verified in this study (i) by comparing the size distribution curves of airborne particles during deposition and during resuspension, (ii) by examination of the individual size distributions for each of the three particle sizes investigated, and (iii) by calculating the resuspended fraction (RF) of initially deposited particles, as determined via Neutron Activation Analysis (NAA).

A comparison of the size distribution curves for deposited and resuspended particles revealed that during resuspension, the highest peak in concentration occurred at $\sim 10 \mu\text{m}$, whereas the highest concentration peak occurred at $\sim 3 \mu\text{m}$ during deposition. When the three distributions were individually analysed, it revealed a shift towards a higher Mass Median Aerodynamic Diameter (MMAD) for the resuspended distribution.

This was confirmed via NAA, which revealed that the percentage of particles resuspended increased with increasing particle size; during high physical activity, an average of 27 ± 9 % of $3 \mu\text{m}$ particles, 30 ± 6 % of $5 \mu\text{m}$ particles, and 34 ± 5 % of $10 \mu\text{m}$ particles resuspended. Additionally, it was revealed that following the collective resuspension of 3, 5 and $10 \mu\text{m}$ sized particles, the larger sized particles were found at their highest concentrations at head height (in comparison to ankle or waist height). This has consequences for a person's potential inhalation exposure; larger particles are less likely to penetrate the lower airways of the lungs, and therefore resuspended aerosol particles do not pose a major threat for increased inhalation exposure.

1 Introduction

Airborne hazardous particles are of major concern in today's society. These potentially dangerous particles include biological aerosol (for example by terrorist release), infectious diseases (transmission in hospitals) and radioactive particles. A major accident at a nuclear power plant (e.g. Fukushima) or an act of terrorism involving large quantities of radioactive material could result in this hazardous material being carried long distances in the atmosphere and affecting populations hundreds of miles away from the source, as was noted by Mustonen (2009) in relation to the spread of radioactive materials over much of Europe following the Chernobyl accident. A country's horticulture and livestock can become contaminated, which exposes the population to contamination by harvesting and ingestion. Inhabitants could also become exposed to the airborne particles through the process of deposition on their hair, skin and clothing (Andersson et al., 2004 and Fogh et al., 1999). Andersson et al. (2002) assumed that on average, 85 % of the human body is covered with clothes.

While deposition and other primary exposure routes have been extensively studied e.g. Lai and Nazaroff (2000 and 2005), Thatcher et al. (2002), less attention has been given to the subsequent fate of deposited particles. Unless a contaminated person then remains perfectly still, they will engage in some degree of human physical activity. This movement will cause particles to resuspend from their clothing and become airborne and thus there is a potential for secondary exposure.

The size of resuspended particles will have implications for a person's exposure type and severity. While the size distribution of hazardous aerosols in the environment is highly variable, in terms of human exposure, the risk varies with particle size. There are many routes for human exposure to hazardous particles including ingestion, inhalation and absorption through the skin. The size of the particles associated with these exposure routes varies. Particles of almost any size can be inhaled but will reach different parts of the lung, depending on their aerodynamic diameter. Particles greater than 10 μm in diameter cannot reach the alveolar region (the unciliated airways) of the lungs and particles greater than 30 μm will not penetrate the respiratory region past the larynx (Cherrie et al., 2010). For contamination deposited onto skin, particles greater than 10 μm cannot penetrate the skin, particles of 3 – 10 μm may reach deeper layers of the skin through the hair follicles but particles less than 3 μm can diffuse through the stratum

corneum to be incorporated into the body's blood supply (Shekunov et al., 2007 and Williams, 2003). These smaller particles ($< 3 \mu\text{m}$) are therefore of high exposure risk as they cannot be safely removed if they have penetrated through the skin. As particle size determines the fate of particles on/in the human body which thus affects a person's exposure risk, it is necessary to fully understand the size distribution of resuspended aerosol particles when determining secondary exposure.

For a surface-residing particle to be resuspended, the lift forces acting on the particle must exceed the forces of attraction between the particle and its residing surface (Hinds, 1999). Particle size affects the forces acting on a particle, as the larger the diameter of the particle, the greater the particle surface area which is in contact with the residing surface and hence a greater lift force is applied to the larger particle. The 'effective area' is the area of contact between the particle and the surface on which lift forces can act (Nicholson, 2009). Increasing sizes of a particular species of particle will have an increase on the particles' effective area and hence, the potential for resuspension is likely to increase with increasing particle size. This will be the case until particle size reaches a threshold diameter at which point the gravitational force will be the dominant force and particles will be restricted from resuspending fully (Hu et al., accessed November 2011). This threshold diameter below which particles can be fully resuspended into the atmosphere (at normal wind speeds) is at approximately $100 \mu\text{m}$ (Nicholson, 2009).

Earlier studies have reported a particle size dependency of resuspended material observed indoors during human activity. Ferro et al. (2004) found that, of the volume of resuspended house dust in a home due to human activity, most of the resuspended particle mass was $> 5 \mu\text{m}$ and submicron particles accounted for less than 1 % of the indoor total suspended particle (TSP) volume. This agrees with the findings of Thatcher and Layton (1995) who concluded that with normal physical activity in a family home, resuspension rate increased with increasing particle size (for a particle size range of $0.3 - 25+ \mu\text{m}$). Abt et al. (2000) concluded that particle emission rates significantly increased (for $0.7 - 10 \mu\text{m}$ particles) with increasing particle size during cleaning and indoor work, due to the resuspension of particles greater than $1 \mu\text{m}$. However these studies all quantified the resuspension of particles due to human physical activity based on a general change in airborne particle concentration, which was assumed to be due to resuspension, but was primarily from floors, and not specifically from a contaminated

person's clothing. Furthermore, these studies do not include a comparison of the size distributions of tracer particles during deposition to that during resuspension. Although Andersson et al. (2004) does compare the deposited and resuspended size distributions of 0.7 and 2.5 μm , indium and dysprosium labelled particles, as with previously cited papers, the particles were resuspended due to vacuuming the floors in a contaminated room and not from a contaminated person's clothing.

The aim of the present work is to quantify the size distribution of particles which resuspend from clothing surfaces during human physical activity and to compare the resuspended particle size distribution to the size distribution of the originally deposited particles.

2 Materials and Methods

A full description of the materials and methods for this work are described in detail in McDonagh and Byrne (2013), but are summarised as follows.

Silica particles of 3, 5 and 10 μm in diameter were respectively labelled with the rare earth metals Europium (EuCl_3), Dysprosium (DyCl_3) and Indium (InCl_3). The three particle sizes were mixed together and collectively aerosolised within a 2.25 m^3 aluminium deposition chamber, using a dry powder particle generator (Palas RBG-1000) and an array of 12 x 33 KBq Am241 radioactive sources, to neutralise any charge acquired by the particles during generation. The particles were allowed to deposit onto clothing samples located on the chamber floor. The clothing types included cotton, polyester, fleece and denim (the significance of the clothing type on particle resuspension is discussed in McDonagh and Byrne (2013)). Large contaminated samples of the clothing were attached to the chest (20 x 20 cm), arms (15 x 30 cm) and legs (25 x 40 cm) of a volunteer. The large samples each had two smaller samples of the same material type pinned onto them and those smaller samples (ranging from 1 x 4 cm to 2 x 5 cm, depending on the material type) were removed for analysis, one each before and after the resuspension event, to determine the proportion of the deposited mass which became resuspended – the Resuspended Fraction (RF).

The volunteer wore a clean room suit (Macrobond hooded coverall from Caulfield Industrial) - chosen for its non-linting and anti-static properties, booties (Polylatex

shoecover 4 g from Caulfield Industrial), gloves (powder free purple nitrile) and a mask (8835 FFP3D disposable mask from Anderco Safety). The volunteer had the contaminated clothing attached to them by an assistant and entered the resuspension chamber - a purpose-built 15 m³ plywood test room. The volunteer engaged in one of two pre-defined physical activities: Low physical activity - walking at a pre-defined rate for 20 minutes, or high physical activity – Irish dancing to a Reel (Wikipedia, accessed Nov 2011) with added swinging of the arms for 10 minutes (chosen to facilitate the accurate reproduction of movement in each repeated experiment). The sampling equipment inside the chamber included: two Aerodynamic Particle Sizer's (APS)'s, one at ankle height (15 cm from the floor) and the other at waist height (90 cm from the floor); two active open-face filters (at 5 L/min), one at ankle height (15 cm from the floor) and the second at head height (150 cm from the floor); three passive filters fixed to the chamber wall at ankle, waist and head height. The filter papers used in each case were Whatman 542 hardened ashless filter papers of 55 mm diameter, chosen because of their suitability for Neutron Activation Analysis (NAA). In the centre of the chamber floor was a grounded metal plate upon which the volunteer stood while performing the experiment. This was to ensure that any static charge generated by the movement of the volunteers' feet, would not influence the behaviour of the resuspended particles.

Three analysis techniques were used. The air within the deposition and resuspension chambers was continuously monitored using Aerodynamic Particle Sizer's (APS) [Model 3321 by Trust Science Innovation (TSI)]. The APS measures the concentration and size of airborne particles in the range of 0.5 to 20 µm in aerodynamic diameter, in real time. The second analysis technique employed was Neutron Activation Analysis (NAA). This allows for the specific detection of the three labelled particles on the clothing material or filter, with a detection limit of ~ 1 pg. The third was Scanning Electron Microscopy (SEM) imagery.

3 Results and Discussion

3.1 Comparison of the Size Distributions of Deposited and Resuspended Particles

Following the experimental procedure as described in section 2, all three particle sizes were mixed together and collectively deposited on to the clothing samples. An APS sampled the chamber air every second during both deposition of the particles onto the clothing samples and resuspension of the particles from the clothing samples.

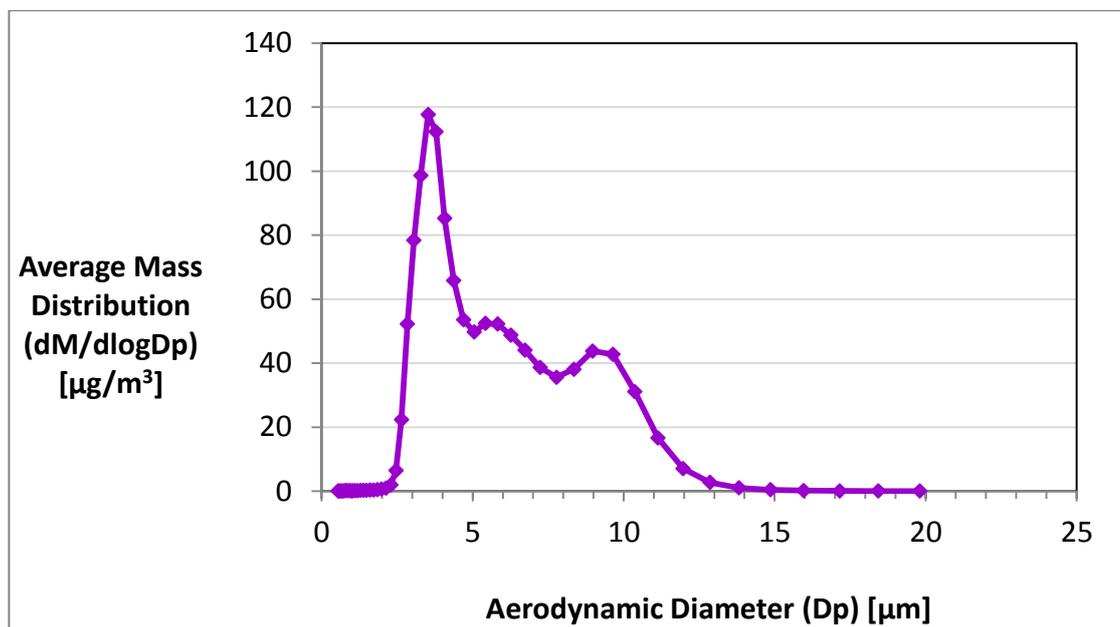


Figure 1. Average mass distribution of airborne particles in deposition chamber during deposition

Figure 1 shows the average airborne particle distribution, in terms of mass, as a function of particle diameter, as seen by an APS in the deposition chamber during deposition of all three particle sizes mixed together. It is clear that there is a mode at approximately 3, 5 and 10 µm. Although the particles injected into the chamber are monodisperse, they each have a size distribution about their Mass Median Aerodynamic Diameter (MMAD) of approximately 3, 5 and 10 µm. These individual distributions overlap but their peak is evident and the remainder of their distributions can be estimated; this will be discussed further in section 3.1.2. For the remainder of this paper, when discussing particle size, the three individual size distributions will be referred to by their concentration peak (or MMAD) of approximately 3, 5 or 10 µm.

Figure 1 indicates that there were considerably more 3 μm sized particles in the deposition chamber than 5 or 10 μm . This may be due to the method used to mix the three particle sizes together before loading into the generator; the three powder samples were simply poured into a mixing container in similar quantities by eye. If the amount by volume of each particle size added was similar, there would be more of the smaller particles than larger particles in the same volume of particles. Any difference in the number of each particle size deposited is not significant, as the resuspended fraction is based on the quantity resuspended as a function of the quantity deposited.

When the size distributions of deposited and resuspended particles are compared, as shown in figure 2, two major differences can be observed. Firstly, the general shape of the size distribution curves for deposited and resuspended particles is markedly different (for all three sized particles mixed together). Secondly, the Mass Median Aerodynamic Diameter (MMAD) of each of the three individual size distributions changes when the deposited particles became resuspended. These differences will be discussed in the next sub-sections.

3.1.1 Change in shape of size distribution curve between deposition and resuspension

In this section, the difference in the general shape of the size distribution curve, between the deposition and resuspension events is examined. For the purposes of this discussion, the resuspension data chosen derives from the case of APS samples taken at waist level during resuspension from denim and under high physical activity conditions.

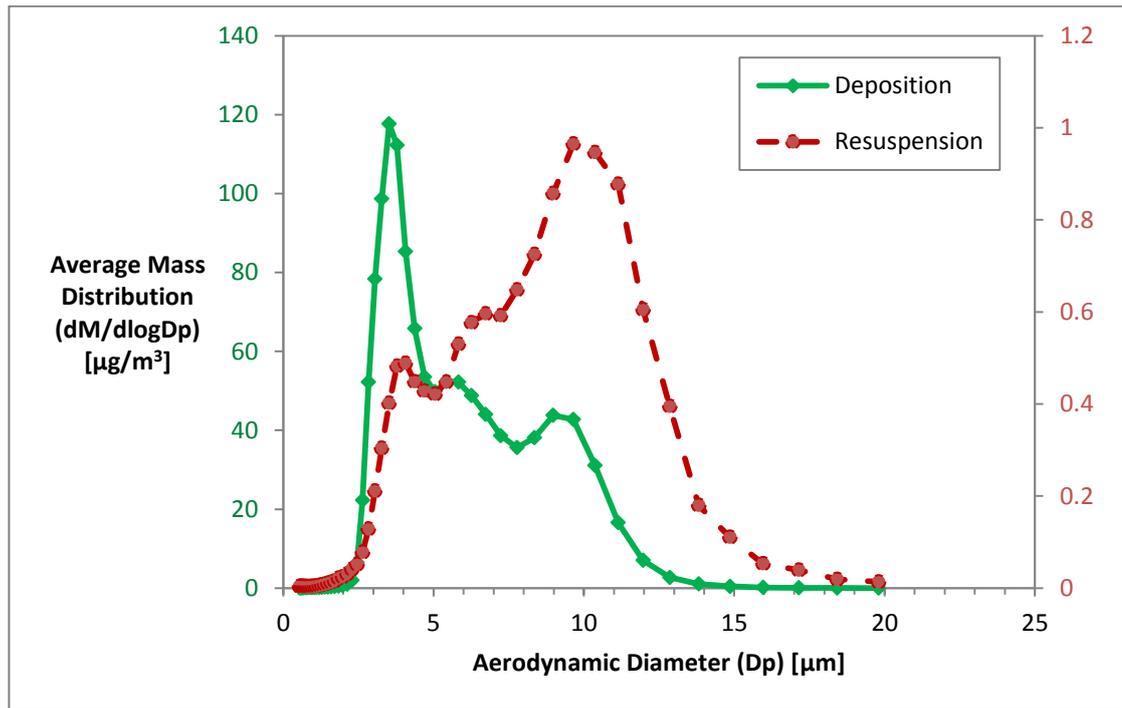


Figure 2. Size distribution of airborne particles in deposition and resuspension chambers to/from denim

In figure 2, the solid green curve represents the airborne particle mass distribution in the deposition chamber, and is shown on the primary y-axis. The dashed red curve is the airborne particle mass distribution during resuspension and is shown on the secondary y-axis.

There is a difference of approximately two orders of magnitude between the concentration of particles deposited and resuspended. This is expected as one would assume that not all deposited particles will resuspend and be detected by the APS. The most significant result from these data is that there is a higher concentration of larger sized particles airborne during resuspension than during deposition. The size distribution curve during deposition (in green) shows its highest mass concentration at approximately 3 µm, with a second smaller peak approximately 5 µm and the smallest peak at approximately 10 µm. These data are summarised in table 1. This illustrates that there are more small particles deposited to the denim than there are large particles, in terms of both mass and number concentrations. Conversely, the 3 µm mass concentration peak on the resuspension curve (shown in red) is the smallest, the second peak is at approximately 5 µm and the third and largest peak occurs at approximately 10 µm. This indicates that more of the larger particles have resuspended from denim than

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the small particles, even though there were more of the smaller particles available for resuspension. However, in terms of number concentration during resuspension, the largest peak occurs at approximately 3 μm and the smallest at 10 μm , which shows an opposite trend to the mass concentration data. However, it is the change in concentration during deposition relative to during resuspension which is important i.e. how many of those particles of a particular size which were available for resuspension, actually resuspended. This value is the same whether one considers the mass or number concentration data, as can be seen in table 1.

	Peak concentration about each distribution:					
	3 μm		5 μm		10 μm	
	Deposition	Resuspension	Deposition	Resuspension	Deposition	Resuspension
Mass Conc. ($\mu\text{g m}^{-3}$)	117.7	0.489	52.41	0.596	43.77	0.965
Number Conc. (# cm^{-3})	8290	34.4	800.8	9.11	83.59	1.84
Percentage change in conc.	- 99.6 %		- 98.8 %		- 97.8 %	

Table 1. Peak concentration in terms of number and mass, about each of the three size distributions, within the overall deposition and resuspension size distributions

The percentage difference in airborne mass concentration between deposition and resuspension, at each of the three particle sizes, is a percentage decrease in each case, as the overall mass concentration during deposition is approximately 100 times greater than during resuspension. The percentage decrease at 3 μm is 99.6 %, at 5 μm is 98.8 % and at 10 μm is 97.8 %, showing a decrease in percentage difference as particle diameter increases. This verifies that more of the larger sized particles are resuspending than are the smaller particles, when comparing the size distribution curve during resuspension to that during deposition.

While the authors of this paper have not found published papers directly comparing size distributions of deposited and resuspended particles from clothing due to human physical activity, Raunemaa et al. (1989) noted that there is a difference between the particle size distribution for deposition and re-emission and that the shift in size is towards the larger sizes. Furthermore, the conclusion that larger particles are easier to resuspend has been widely observed both theoretically and experimentally. Thatcher and Layton (1995) examined the increase in particle concentration indoors during various human activities, for four different particle size ranges. They concluded that particles between 0.5 and 1 μm in diameter were essentially non-resuspendable, whereas particles between 10 and 25 μm showed approximately a 6-fold increase in airborne particle concentration above background levels, with 2 minutes of continuous walking and sitting by one person in the living area of a house. Abt et al. (2000) concluded that particle emission rates significantly increased (for 0.7 – 10 μm particles) with increasing particle size during cleaning and indoor work, due to the resuspension of particles greater than 1 μm . Andersson et al. (2004) investigated the resuspension of particles from 0.4 to 7 μm from various surfaces (wood, plastic and wool), by dropping a weight onto the contaminated surface. They found that in the case of all three surfaces, there is a shift towards a larger count median diameter for the resuspended particles relative to the deposited particles. While the findings of these papers agree with the findings reported in the current work, these studies were quantifying the resuspension of particles based on a general change in airborne particle concentration, which was assumed to be due to resuspension but primarily from floors, and not specifically from a physically active contaminated person's clothing.

3.1.2 Mass Median Aerodynamic Diameter (MMAD) shift between deposition and resuspension, for each size spectrum

The Mass Median Aerodynamic Diameter (MMAD) for each particle distribution, during deposition and resuspension is summarised in table 2. The data are averaged over all material types and for airborne mass concentrations as measured during high physical activity.

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	Median particle diameter (in μm) of each distribution:					
	3 μm		5 μm		10 μm	
	Deposition	Resuspension	Deposition	Resuspension	Deposition	Resuspension
MMAD	3.52	4.00	5.43	6.50	8.98	9.83
Percentage change in MMAD	+ 0.12 %		+ 0.16 %		+ 0.09 %	

Table 2. Mass Median Aerodynamic Diameter (MMAD) of each of the three particle size distributions, during both deposition and resuspension

As can be seen from table 2, for each of the three size distributions, for both mass and number concentrations, the MMAD of particles resuspended is greater than that of particles deposited. For the 3 μm distribution, the average MMAD of deposited particles is 3.52 μm and of resuspended particles is 4.00 μm , a 13.6 % increase in the peak diameter of particles resuspended in comparison to those deposited. For the 5 μm distribution, the average MMAD of deposited particles is 5.43 μm and of resuspended particles is 6.50 μm (an increase of 19.7 %) and for the 10 μm distribution, the average MMAD of deposited particles is 8.98 μm and of resuspended particles is 9.83 μm , an increase of 0.85 μm .

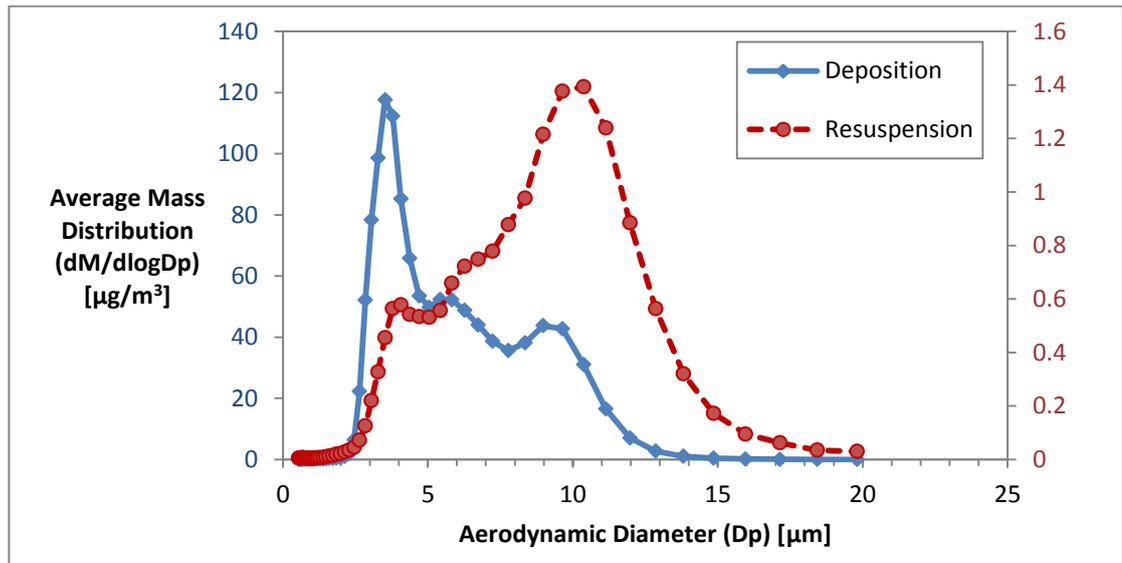


Figure 3. Average size distribution of airborne particles during deposition and resuspension at high physical activity

Figure 3 is a graphical representation of the mass concentration data from table 2 and confirms the findings in section 3.1.1; that the amount of particles resuspended increases with increasing particle size, and within the distribution of deposited particles of a particular size, the peak size of particles resuspended shifts slightly to the right or to a larger diameter, in comparison to the peak size of those deposited. So within a distribution, the larger particles resuspend more and thus the MMAD of the distribution is higher. This statement can be confirmed by separating the size distribution curves for each of the three particle sizes and forecasting their curve to complete the distributions from where the three curves overlap. This is done in figure 4, where the distribution for the 3 µm is separated from the full distribution and therefore its final point (before the distribution overlaps with the distribution about 5 µm) is at ~ 4.1 µm. The curve is then forecast forward to complete its estimated distribution. The curve is forecast using the knowledge that monodisperse airborne particles of this size range and generated by this method, are generally expected to follow a Gaussian distribution (Colbeck, 1998). Using the known portion of the distribution about the 3 µm peak, the remainder is estimated by hand, using a Gaussian curve with a sigma (standard deviation) of 1 (as this provided the best fit to the data) and is represented graphically in figure 4. The forecasted portion of the dotted blue deposition curve is shown by a solid blue line and the forecast portion of the dashed red resuspension curve is shown by a solid red line.

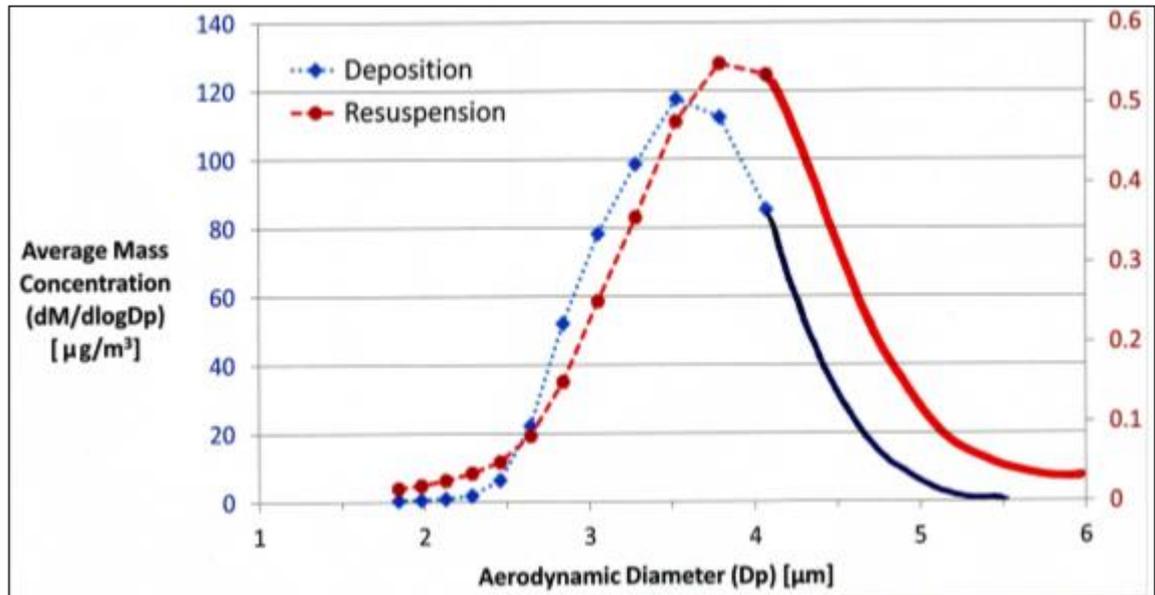


Figure 4. Forecast size distribution of airborne particles in deposition and resuspension chambers, to/from cotton (focus on 3 μm)

Figure 4 shows that the resuspension curve has shifted to the right in comparison to the deposition curve. This confirms that the mean particle diameter of resuspended particles was greater than that of the deposited particles, which implies that, of the deposited particles, those with a larger diameter were more likely to resuspend.

A shift in MMAD was also noted by Andersson et al. (2004) who examined the mean diameter of tracer particles released into a test room and compared it to the mean diameter of those deposited particles which became airborne after the room surfaces were agitated with a filter-less vacuum cleaner. Andersson et al. (2004) released tracer particles with two different size distributions and compared their mean diameter before and after resuspension events. Andersson concluded that the particle distribution with a mean diameter of 0.7 μm before agitation had a mean diameter of 2.8 μm after agitation, which occurred 24 hours after the deposition. However the particle distribution with a mean diameter of 2.5 μm before agitation showed no significant change in size distribution after any of the resuspension events. In contrast, the results presented in this paper show an increase in the MMAD following resuspension for all three particle sizes examined (3, 5 and 10 μm).

The greater tendency for large particles to resuspend than smaller particles, as evidenced in the current work, can be explained by examining the forces acting on a surface-residing particle. It has been extensively studied and generally understood that

the magnitude of adhesive forces is directly proportional to the first power of particle diameter, but removal forces are proportional to the second (gravitational, vibrational and centrifugal forces) or third power (air currents) of diameter (Hinds, 1999). Thus particles of increasing size are easier to resuspend from a surface. The knowledge that particles with large diameters are more likely to resuspend, has implications for estimating the impact on a persons' exposure to hazardous particles. In terms of inhalation exposure, less than 2 % of large particles ($> 10 \mu\text{m}$) will pass the nose and upper airways of the lungs and thus it can be concluded that as resuspended particles are of a relatively large size, their significance for respiratory exposure is less than by other exposure routes (Byrne, 2009). Also larger particles ($> 3 \mu\text{m}$) are less likely to penetrate a person's skin and thus will not be readily absorbed into the body (Shekunov et al., 2007).

3.2 Relationship between Particle Size and the Resuspended Fraction (RF)

The purpose of this section is to examine the resuspended fraction (RF) data for each particle size, at low and high physical activity levels. The data are averaged over all material types and body locations. Low physical activity consisted of walking for 20 minutes and high physical activity was representative of running and simulated by modified Irish dancing to an Irish Reel for 10 minutes, as described in section 2.

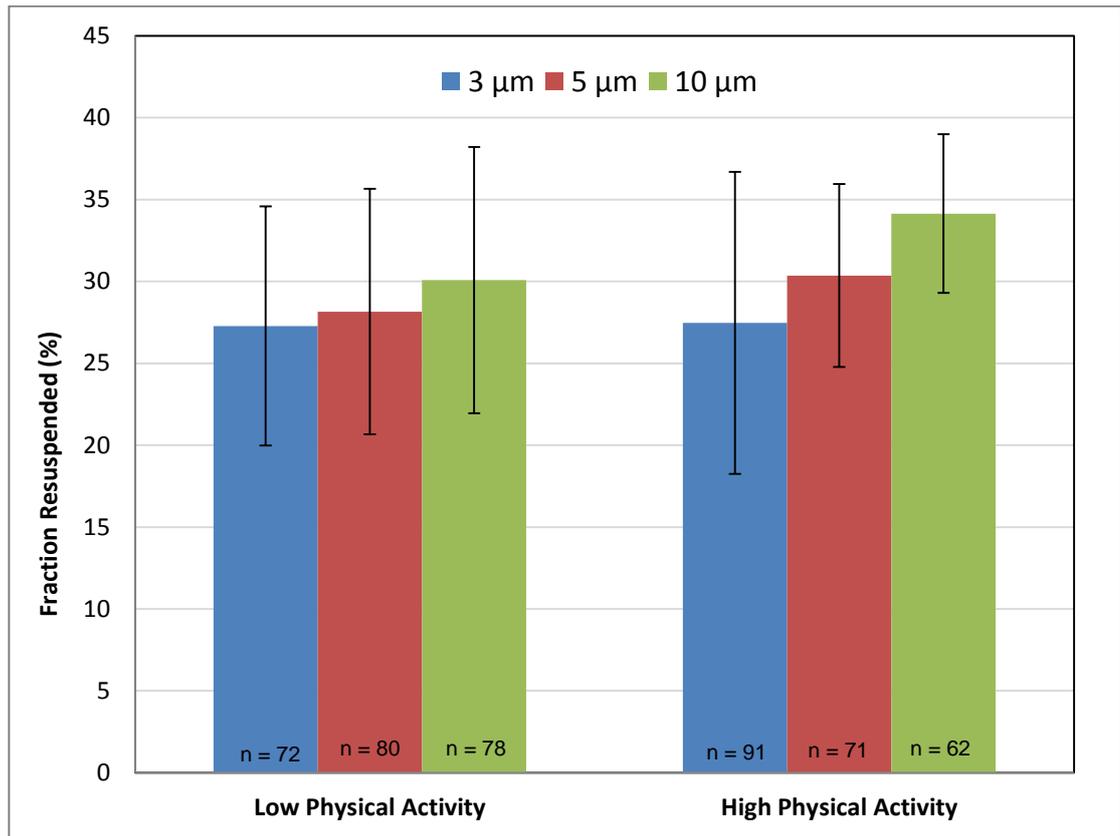


Figure 5. Percentage of the original deposit which has resuspended from all surface types, at low and high physical activity

Figure 5 shows the fraction of each particle size which becomes resuspended due to low and high physical activity. The height of the bars indicates the fraction of originally deposited particles of a specific size which have been resuspended. The bar colours represent the different particle distributions with 3 μm shown in blue, 5 μm in red and 10 μm in green. The number of separate experimental values that were averaged is indicated on each bar. The error bars indicate one standard deviation above and below the average value in each data set.

These data show that the fraction of particles which become resuspended from the clothing of a person engaged in low physical activity, ranges from 8 to 52 % and with an average RF across all sized particles of 28 ± 8 %. The average RF from all body locations and material types for the 3 μm particles is 27 ± 7 %. For the 5 μm particles is 28 ± 7 % and for particles of 10 μm in size, an average of 30 ± 8 % of the originally deposited particles have resuspended.

During high physical activity, between 3 and 67 % of particles formerly deposited on various clothing types was found to resuspend, with an average RF value across all particle sizes of 30 ± 7 %. On average, 27 ± 9 % of 3 μm particles, 30 ± 6 % of 5 μm particles and 34 ± 5 % of 10 μm particles resuspended. As with the low physical activity, these data suggest that the fraction of particles resuspended increases as particle size increases. However, statistically analysing the data using a two-tailed t-test for samples of equal variance indicates that the differences in the RF between sizes for each physical activity level is not significant for $p = 0.05$. This apparent lack of difference in RF between the three sized particles is only seen on the samples analysed by NAA (figure 5). Analysing the airborne mass concentration of particles in the air surrounding a person engaging in high physical activity as measured by an APS (figures 2 and 3), shows there is a significant difference in the distributions about each particle size, for a significance level of 0.05. This apparent contradiction is due to the capturing of different features of the resuspension process by the two different analytical techniques, as discussed below.

The APS data shows the airborne mass concentration as a function of particle size and thus measures all particles that are airborne, regardless of their origin. NAA data are used to determine the resuspended fraction (RF), which is calculated by measuring the difference in mass concentration of particles deposited on the material sample before and remaining on the material sample after the period of resuspension, as a percentage of the mass concentration deposited on the material sample before resuspension. Hence the concentration of particles remaining on a sample after resuspension is the key variable in determining the RF from that sample. However, this value may be higher than expected due to an unquantified number of particles remaining on that sample after the resuspension event, caused mainly by three processes. The processes are discussed in detail in McDonagh and Byrne (2013) and are: 1. Impaction of resuspended particles back onto the material sample, as it passes through the particle laden air stream. 2. Enhanced deposition due to both increased air velocities around the moving person and the rate of displacements of the body parts (and hence the material samples) themselves. 3. Penetration of resuspended particles deep into the weave of the material. Because of the particles' increased velocities when they redeposit, they will have an increased kinetic energy and hence may penetrate deeper into the weave pattern of the material and therefore be more difficult to re-resuspend.

As a consequence of the above three events occurring during resuspension, the mass of particles calculated to be on the clothing sample after the period of resuspension may be higher than would be the case if all particles which initially resuspended were removed and not available to re-deposit. Therefore a lower RF than is actually the case is calculated but it should be noted that this RF is representative of a real-life scenario where impaction and secondary resuspension would be continuously occurring.

3.3 Relationship Between Particle Size and the Height Reached by Resuspended Particles

Passive and active filters were operational during all resuspension experiments. There were 3 passive filters at heights of 15, 90 and 150 cm from the chamber floor (representative of ankle, waist and head heights) and 120 cm from the physically-active contaminated person who was the source of particles. Also, two active filters were positioned at heights of 15 and 150 cm from the chamber floor (ankle and head heights) and 75 cm from the source, in open-face filter holders through which air was drawn at approximately 5 L min^{-1} . The two active filters were 45 cm closer to the contaminated person than the passive filters. After each experimental run, all five filters were analysed by NAA to determine the concentration of each particle size on each filter.

The following figures show the concentration of each element – Europium (Eu), Dysprosium (Dy) and Indium (In) – detected by NAA, on filters placed at specific heights from the resuspension chamber floor. It is the average concentration over all material types and physical activity levels, on passive and active filters. The concentration values are in units of $\mu\text{g kg}^{-1}$ which is the mass of the element detected in micro-grams divided by the mass of the sample (filter paper plus particles) in kilograms i.e. the concentration in parts per trillion. This ensures that any difference in the size (and hence mass) of the samples is taken into account. The Eu, Dy and In are labels chemically bonded onto particles of diameter 3, 5 and 10 μm respectively and hence the mass of the element detected on the filter paper, is representative of the mass of that sized particle on the filter paper.

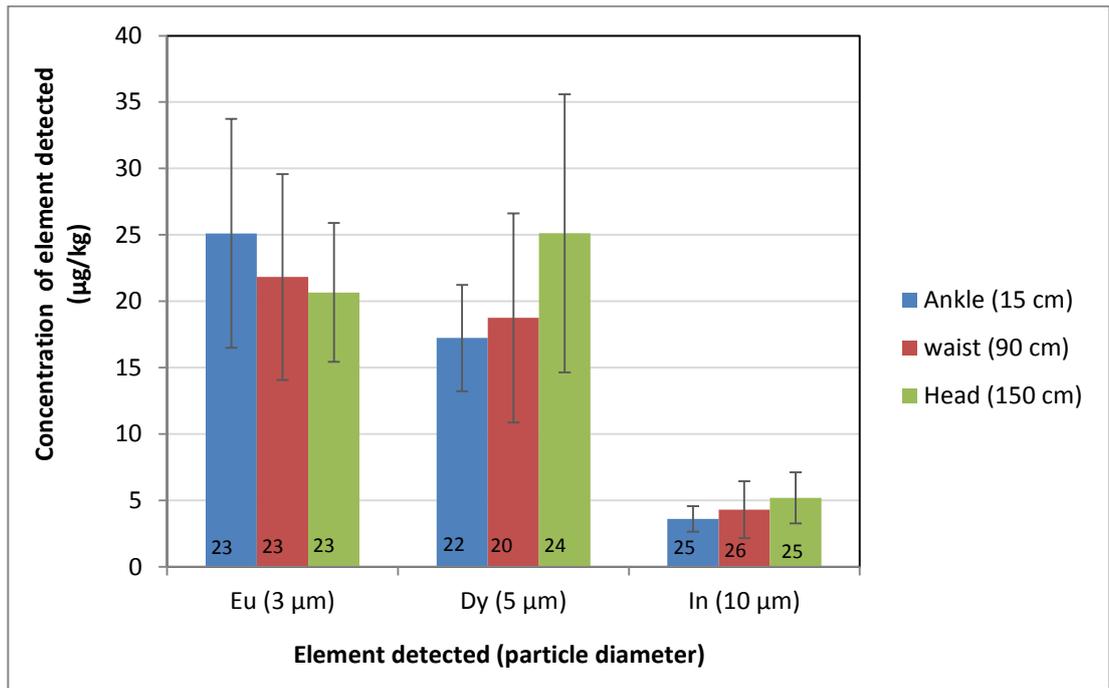


Figure 6. Concentration of 3, 5 and 10 µm particles as detected on Passive filters at various heights, from all material types and physical activity levels

The above graph shows the concentration of each element (and thus mass concentration of each particle size) detected on the passive filters at ankle, waist and head height. The concentration of particles detected on the filters at ankle height is represented by the blue bars, at waist height is in red and at head height is in green. The value shown at the bottom of each bar indicates the number of experimental values averaged. The error bars show one standard deviation above and below the average value. The values are summarised in table 3.

By examining the distribution of 3 µm sized particles, across the three passive filter heights, it can be observed that the mass concentration of the particles captured decreases as the filter height above the ground increases, with an $18 \pm 5\%$ decrease in the mass of resuspended 3 µm particles between filters at ankle and head height.

A different trend is observed for the larger particles. Specifically for 5 µm and 10 µm particles; as the filter height above the ground increases, the mass concentration of particles captured also increases, with $46 \pm 15\%$ increase in the mass of resuspended particles between ankle and head height, for particles of 5 µm and a corresponding increase of $44 \pm 14\%$ for 10 µm.

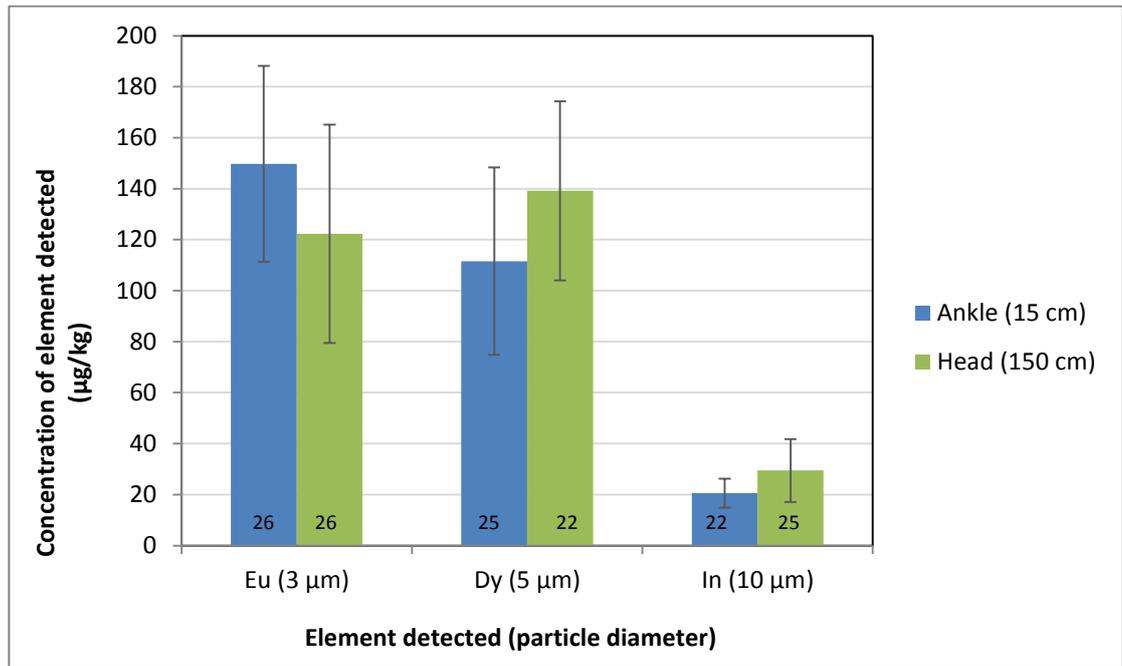


Figure 7. Concentration of 3, 5 and 10 µm particles as detected on Active filters at various heights, from all material types and physical activity levels

Figure 7 shows the concentration of each element (and thus mass concentration of each sized particle) detected on the active filters at ankle and head height. The concentration of particles detected on the filters at ankle height is represented by the blue bars and at head height is in green. The number at the bottom of each bar indicates the number of experimental values averaged. The error bars show one standard deviation above and below the average value. The values are summarised in table 3.

For the 3 µm particles, as the filter height above the ground increases, the mass concentration of particles captured decreases, with an $18 \pm 5 \%$ decrease in the mass of resuspended 3 µm particles which deposited onto filters at ankle and head height. However, for 5 and 10 µm particles; as the filter height above the ground increases, the mass concentration of particles captured increases, with a $25 \pm 7 \%$ and $43 \pm 14 \%$ increase in the mass of resuspended particles between ankle and head height for particles of 5 and 10 µm, respectively.

A STUDY OF THE SIZE DISTRIBUTION OF AEROSOL PARTICLES
RESUSPENDED FROM CLOTHING SURFACES

	Passive Filters			Active Filters		
	3 μm	5 μm	10 μm	3 μm	5 μm	10 μm
Ankle (15 cm)	25.1 ± 8.6	17.2 ± 4.0	3.6 ± 1.0	149.7 ± 38.5	111.6 ± 36.7	20.6 ± 5.7
Waist (90 cm)	21.8 ± 7.8	18.7 ± 7.9	4.3 ± 2.1	-	-	-
Head (150 cm)	20.7 ± 5.2	25.1 ± 10.5	5.2 ± 1.9	122.3 ± 42.8	139.2 ± 35.1	29.5 ± 12.3
% change in conc. between ankle and head	- 18 $\pm 5 \%$	+ 46 $\pm 15 \%$	+ 44 $\pm 14 \%$	- 18 $\pm 5 \%$	+ 25 $\pm 7 \%$	+ 43 $\pm 14 \%$

Table 3. Summary of mass concentration (in $\mu\text{g}/\text{kg}$) of each particle size \pm S.D., as captured by passive and active filters, at specific heights from the floor

In summary, there are less 3 μm particles at head than at ankle height but the opposite is true for particles of 5 and 10 μm in diameter where a higher concentration is seen at head height. This may be due to larger particles being subjected to greater lift forces from their residing surface (Hinds, 1999) due to their larger effective area and thus being resuspended higher into the air. Therefore the larger particles will be captured by the higher filters before they deposit to the chamber floor. Correspondingly, the smaller particle size of 3 μm will be subjected to a smaller lift force and will therefore fall to the lower filter for capture. This phenomenon might not have been seen if the filters were located further from the source of particles as, once they have been resuspended from the source, the larger particles would deposit to the ground quickly due to their large mass and therefore not reach the head height filters if they were located further from the source.

While there were far more particles (a 6.5 fold difference) captured by the active filters ($286.5 \mu\text{g kg}^{-1}$) than by the passive filters ($44.3 \mu\text{g kg}^{-1}$), the percentage change in

particle concentration between ankle and head height is almost identical for the 3 and 10 μm particles, regardless of filter capture type (table 3). Between the ankle and head regions, there is an $18 \pm 5\%$ decrease in the concentration of 3 μm particles for both passive and active filters, with a $44 \pm 14\%$ and $43 \pm 14\%$ increase in the case of 10 μm particles on passive and active filters respectively. For the 5 μm particles both filter types show a percentage increase in particle concentration between the ankle and head heights. While these percentage differences have values of $+46 \pm 15\%$ on passive filters and $+25 \pm 7\%$ on active filters, they are not significantly different (according to a t-test).

This observation of an increase in the concentration of larger particles at head height relative to other locations has implications for the potential inhalation exposure resulting from resuspended hazardous particles, as particles with a diameter of 10 μm or greater are less likely ($< 2\%$) to penetrate to the lower airways of the lungs. Also, if the smaller sized resuspended particles, which are more hazardous in terms of inhalation exposure, are found in higher concentrations at ankle height, they are likely to deposit to the ground and thus could be more easily removed from the environment by cleaning etc. They therefore do not pose a major threat for increased inhalation exposure. In summary; these results indicate that while resuspension will significantly increase airborne particle concentrations, which has implications for general exposure risk, current findings regarding the size of the resuspended particles and the height that the various sizes can reach indicates that resuspended material has a lesser consequence in respect of inhalation exposure, than it has by other routes.

4 Conclusions

When the size distributions of deposited and resuspended particles were compared, two major differences were observed. Firstly, the general shape of the size distribution curves for deposited and resuspended particles was markedly different (for all three sized particles mixed together). There was a difference of approximately two orders of magnitude between the concentration of particles deposited and resuspended. Also, more of the larger sized particles resuspended than did smaller particles, following comparison of the size distribution curve during resuspension to that during deposition.

Secondly, the Mass Median Aerodynamic Diameter (MMAD) of each of the three individual size distributions changed when the deposited particles became resuspended. The peak size of particles resuspended shifted slightly to the right or to a larger diameter, in comparison to the peak size of those deposited. Furthermore, the entire resuspension curve shifted to the right in comparison to the deposition curve. This confirms that the mean particle diameter of resuspended particles was greater than that of the deposited particles, which implies that, of the deposited particles, those with a larger diameter were more likely to resuspend.

The fraction of deposited particles which resuspended (the resuspended fraction (RF)) from a person's clothing was examined for two different physical activity levels. The RF from the clothing of a person engaged in low physical activity, ranged from 8 to 52 % and with an average RF across all sized particles of 28 ± 8 %. During high physical activity, between 3 and 67 % of particles formerly deposited on various clothing types was found to resuspend, with an average value across all particle sizes of 30 ± 7 %. These percentages are significant in terms of the transport of hazardous aerosol particles in the environment and hence resuspension from contaminated clothing is potentially a large source of secondary exposure to humans.

The relationship between particle size and the height reached by resuspended particles was also examined, as this can have an effect on the type and severity of a person's exposure. By examining the distribution of 3 μm sized particles, across the three filter heights (ankle, waist and head), it was observed that the mass concentration of the particles captured decreased as the filter height above the ground increased, with an 18 ± 5 % decrease in the mass of resuspended 3 μm particles between filters at ankle and head height. The opposite trend was observed for the larger 5 μm and 10 μm particles; as the filter height above the ground increased, the mass concentration of particles captured also increased. For particles of 5 μm , a 46 ± 15 % increase in the mass of resuspended particles between ankle and head height was observed on passive filters and a corresponding increase of 44 ± 14 % for 10 μm particles. Further studies could repeat these experiments but with the sampling locations at varied distances from the source, to examine if filters at a greater distance would exhibit the same trends as revealed here.

In conclusion, the generally accepted theory that larger particles are more likely to resuspend, has been confirmed in this study. The extent of which formerly deposited particles will resuspend from a person's clothing during physical activity, has been quantified for three specific particle sizes, 3, 5 and 10 μm . Future work could extend the scope of this study by including submicron sized particles.

5 Acknowledgements

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Mass Transport of Deposited Aerosol Particles by Surface-to-Surface Contact

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Abstract

The spread of particle-borne contamination by surface-to-surface contact and its implications for exposures within the indoor environment has been observed - largely qualitatively. The present study was conducted with the aim of quantifying the mass transfer efficiency (TE) of deposited aerosol particles when selected soft and hard surfaces come in contact. The surfaces used were 100 % cotton, synthetic fleece, plastic laminate and brass. Contact transfer efficiencies ranging from 2 to 45 % were observed; these are very significant numbers in terms of hazardous aerosol transport in the environment. Other observations include an increase in the mass transferred with increased surface roughness. An increase in the applied pressure between the two surfaces in contact leads to a step change in transfer efficiency, so that two pressure regimes can be identified, with a transition pressure between them that depends on surface type. Time of contact appears to have little to no effect on the mass transfer efficiency for the surfaces studied, while contaminant loading has some effect that is not systematic.

1 Introduction

The spread of deposited aerosol particles from one surface to another via contact transfer is an important factor in assessing the exposure pathways of airborne contaminants. Indoor airborne contaminants will deposit on indoor surfaces; the deposition loss rate depends upon aerosol size, surface type (e.g., hard vs. soft) and orientation (e.g. horizontal or vertical) and flow conditions within the indoor space. Transport and fate of indoor aerosols have been extensively studied both experimentally and theoretically, see, for example Abadie et al. (2001), Byrne et al. (1995), Fogh et al. (1997), Lai and Nazaroff (2000, 2005), Thatcher et al. (2002), and references therein. Less attention has been given to the subsequent behaviour of deposited particles. For example, deposited particles can be spread from one surface to another through the process of contact transfer. The process of surface to surface contact will usually involve human activity. For example a person coming into contact with aerosol contaminated surfaces, such as sitting on a chair or resting their arms on the desk, will transfer a portion of the deposited contamination to their clothing. Subsequent transfer of contamination is also possible. If the person next comes into contact with a 'clean' surface, some fraction of the contamination on their clothing will transfer to the second surface.

Andersson et al. (2004) concluded that when particles of 0.7 and 2.5 μm are deposited on clothing, less than 5 % will penetrate through the clothing. A wide range of fabrics were tested, including thick and thin pure cotton, thin 65 % polyester and 35 % cotton, thick 50 % wool and 50 % acrylic. If very few particles penetrate through the clothing but remain on the surface, then the particles have a potential for resuspension or transfer to other surfaces by contact. Some studies have demonstrated the spreading of contaminant via the process of contact transfer but aerosol mass transfer from one surface to another has been poorly quantified. Clothing has been identified as a significant source of contaminant transfer in the indoor environment. Tovey et al. (1995) state that dust mite allergen found in clothes could come from many sources including allergens picked up when clothing items were laid on furnishings, floors and beds. De Lucca et al. (2000), Liccardi et al. (1998) and D'Amato et al. (1997) indicate that clothing is an important means of distributing cat allergen into cat-free environments.

Cross-contamination by staff uniforms in the medical industry has also been examined. Loh et al. (2000) showed that the sleeves of medical students white coats were more

likely to contain bacterial colony counts than the backs of the coats. This is most likely due to contact transfer of particles from patients themselves, their clothing or bedding to the medical student as the patient is being examined. While Perry et al. (2001) did not directly assess if contamination present on nurses uniforms is transferred to other patients, it was noted that some uniforms which showed a positive level of contamination at the start of a shift, were negative at the end. While the lack of bacterial detection at the end of the shift does not definitively suggest loss of mass due to transfer to other surfaces, it is possible that some fraction of the contamination was transferred from the uniform to the hospital environment.

Mass transfer rates between skin and various other surfaces have been extensively documented. Reed et al. (1975) studied the transmission of Rhinovirus colds through indirect contact and found that the virus could be spread from an infected person to another person via intermediate objects but that it was necessary to determine the efficiency of virus transfer from surface to surface. Brouwer et al. (1999) conducted an experimental study on dermal exposure in the workplace resulting from contact with contaminated sources for input into risk assessment models. This was achieved by investigating the transfer of a fluorescent whitening agent in powder form from a contaminated glass plate to the skin on a hand. The authors did not report the size distribution of the powder particles. Comparison was made among the contaminant loading, increase in area exposed and adherence to the skin after 1 to 12 consecutive contacts. It was concluded the transfer efficiency (TE) was $\leq 2\%$ of the original contaminant on the surface. The time of contact varied from 3 to 30 seconds and contact pressure was kept constant. These authors concluded that contaminant loading on the skin increased nearly linearly with increased contacts, for up to 6 consecutive contacts. There was a decrease in additional loading with consecutive contacts beyond the sixth contact. It was also found that the quantity of contaminant transferred to the skin was dependent on the surface mass contamination. Hubal et al. (2005) investigated the TE of the fluorescent tracer riboflavin between skin and other surfaces (including carpet and laminate) and made similar conclusions to Brouwer et al. (1999), in that surface loading and skin condition were among the most important parameters for characterising residue transfers. It was also concluded that contact time did not significantly affect transfer.

As discussed above, previous studies have demonstrated contaminant transfer between clothing and other surfaces, including skin. However, to the best of the authors' knowledge surface-to-surface mass transfer efficiencies have not been previously

quantified. The aim of this study was to quantify the mass transfer efficiencies of deposited aerosol particles to and from selected hard and soft surfaces typically found in a home or office. Other variables investigated included the applied pressure, contact time and contaminant loading. Because the number of potential surface pairs is quite large, two soft surfaces and two hard surfaces were chosen as examples of the types of surfaces commonly found indoors. The goal of these initial experiments was to determine the range of transfer efficiencies and to provide a preliminary investigation of the influence of several variables on these efficiencies.

2 Methods

The surfaces to be contaminated were first exposed to aerosol particles generated from pure fluorescein powder. Contact was then made between the contaminated surface and a clean surface for a specified length of time and contact pressure. Both the contaminated and receiving surfaces were then analysed using the techniques described below to determine the proportion of the original deposited mass transferred to the receiving surface. The experimental details are described in the next sub-sections.

2.1 Surfaces

Four common surfaces were chosen for these initial experiments, brass, plastic laminate and fabrics of cotton and fleece. The plastic laminate material is typical of that found on indoor surfaces such as counter tops and desk tops. The two soft fabric surfaces chosen were 100 % cotton and a synthetic fleece. Cotton is a popular textile used in numerous clothing items such as t-shirts and denim jeans and in surface coverings such as furniture cushions. Fleece was chosen as it is a synthetic fabric in contrast to the natural cotton and it has a more textured surface than the cotton. Before use all surfaces were prepared as follows: The cloth was prewashed in a standard clothes washing machine using washing soda (in place of laundry detergent) and dried in a standard clothes dryer at low/medium heat. Cotton cloth was ironed to remove major wrinkles. Samples of the washed cloth were subjected to the extraction procedures discussed below to ensure there was no fluorescent background.

The plastic laminate, cotton and fleece cloth pieces measured 15 x 10.5 cm, with a 10.5 x 10.5 cm square marked in their centre to define the area exposed to aerosol deposition.

The brass pieces measured 25 x 10.5 cm, for reasons described in Section 2.2, brass and plastic laminate pieces were pre-treated by first wiping with ethanol, then thoroughly washing in warm water and finally rinsing in an alkali buffer solution. All surfaces were immersed in the buffer solution described in Section 2.4 below to test that no background fluorescence could be extracted after the surface treatment. None of the test surfaces were found to fluoresce within the detection spectrum that would cause interference.

2.2 Aerosol Deposition

The surfaces used in the contact transfer experiments were contaminated in a 0.5 m³ aluminium deposition chamber, where a small 20 W fan is mounted in the back right corner of the chamber, 34.5 cm from the ceiling and facing the opposite front left corner. The fan helps ensure uniform aerosol mixing within the chamber. The airborne particle concentration was monitored both by filters that were later analysed for fluorescence and by a real-time instrument. Two open face filter air samplers were operational during deposition events, one at each side of the chamber and sampling at ~ 6 L min⁻¹. An aerosol size instrument (APS – Aerosol Particle Sizer, TSI Model APS 3321) was positioned under the chamber and sampled through a metal tube of diameter ~ 1 cm, passing through the centre of the 0.56 m² chamber floor and extending 8 cm above the floor. Polydisperse fluorescein particles are injected into the chamber through a disperser mounted inside the chamber near the ceiling. This disperser has radial holes to provide lateral flow and is connected to a jar outside the chamber via a 1 cm diameter Cu tube inserted through the centre of the chamber ceiling. The jar contains the dry fluorescein powder which is aerosolized by air at 245 kPa and a flow rate of 18 L min⁻¹. The injected particles were in the size range of 0.5 – 20 µm diameter with an MMAD (Mass Median Aerodynamic Diameter) of 12.9 ± 3.8 µm, as measured by the APS during deposition.

The chamber was cleaned between experiments. The various surfaces were then placed on the chamber floor – which was first covered by clean paper towels to avoid fluorescein contamination of the bottom side of the deposition surfaces – and surrounded by 25 x 10.5 cm brass plates which were arranged to mask the rectangular cloth pieces into 10.5 x 10.5 cm squares. Eight surface samples were contaminated at a time. Four surface samples were arranged in the back half of the chamber and four in

the front half, along with three pieces of brass in each half of the chamber. The brass and surface samples were interspersed in each half of the chamber as follows: (from left to right) brass sheet – two surface samples – brass sheet – two surface samples – brass sheet. After each deposition ‘run’, the mass loading on each of the six brass sheets was determined, providing a measure of the spatial variation in deposition.

When the surfaces were arranged on the chamber floor, the door was sealed and the APS, air sampling pumps and fan were turned on. Then the air supply to the jar containing the fluorescein powder was turned on until the APS showed an airborne particle number concentration in the chamber of $\sim 1000 \text{ cm}^{-3}$. This typically took ~ 25 seconds. The air supply was then turned off while the fan, APS and air sampling pumps were left in operation until the airborne particle number concentration dropped to $\sim 300 \text{ cm}^{-3}$, which took ~ 30 minutes. A small, 1-cm-diameter hole in the side of the chamber connected the inside of the chamber and the lab room. This hole made it easier to maintain a slightly negative pressure differential between the chamber and the lab room under the two different airflow regimes used during the deposition runs. When only the APS and air sampling pumps were operating, the differential pressure was $\sim -3 \text{ Pa}$, and during the time the aerosol generator was being used, the pressure differential was $\sim 0 \text{ Pa}$. Background aerosol number concentrations in the lab room air were typically $\sim 20 \text{ cm}^{-3}$, quite low compared with the chamber concentrations so there should be no interference from ambient particles.

2.3 Contaminant Transfer Measurements

Following the deposition process, the contaminated surface was carefully transferred from the chamber onto a piece of clean brass (15 x 15 cm) mounted horizontally and held into position using small metal spring clips at the edges of the surface (outside the contaminated area – see figure 1). The configuration of the receiving surface varied depending on the surface type. For the cloth receiving surfaces a brass holder was constructed from a flat rectangle the same dimensions as the cloth pieces. This piece of brass had the two ends of the rectangle turned up at a 90° angle, leaving a 10.5 cm by 10.5 cm flat surface in the middle. The holder was centred on the cloth receiving surface and the two ends of the cloth rectangle were wrapped around these brass sides and attached at the ends with four spring clips (see figure 1 below). This method ensured that the receiving surface area coming into contact with the contaminated surface was

undisturbed by the clips. A 10 x 10 cm wooden block on the inside of this holder helped ensure that the pressure generated by the applied weight was distributed uniformly across the contact surface. As the receiving plastic laminate could not be bent up over the ends of this brass holder, it was simply placed down onto the contaminated surface, taking care to ensure that the 10.5 x 10.5 cm squares in the centre of both surfaces were aligned.

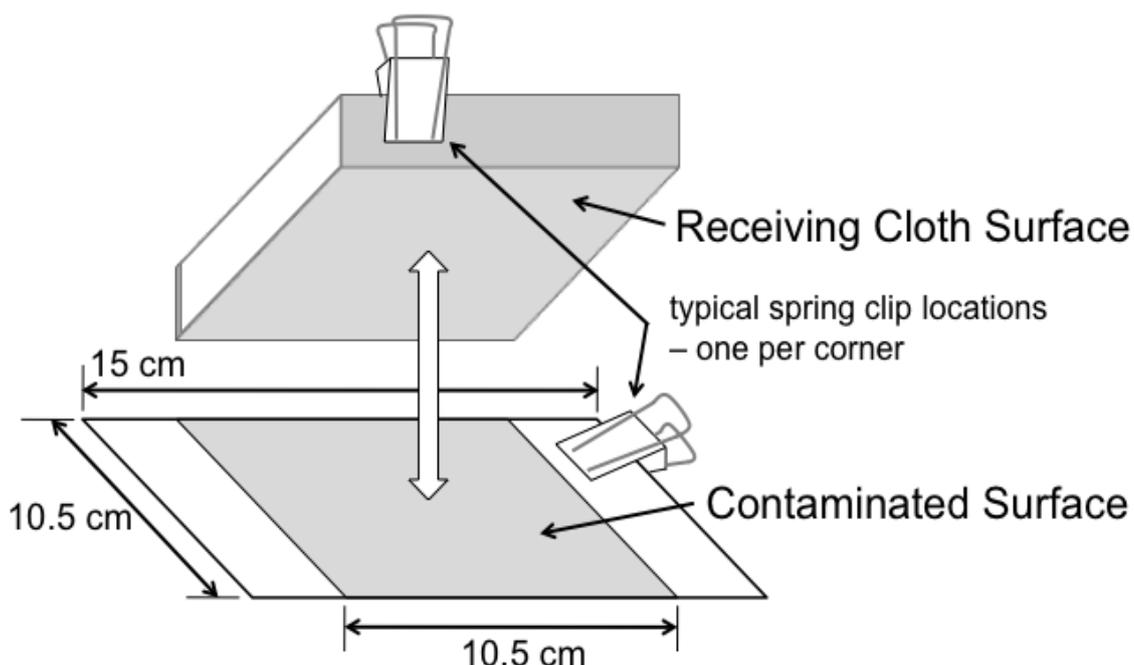


Figure 1. Schematic diagram of the contact transfer method. The contaminated surface is held down to a flat brass plate by a spring clip at each corner. The cloth receiving surface is mounted on a brass frame using spring clips on the upturned sides – one on each corner

The contact area between the upward-facing contaminated and downward-facing receiving surfaces was $\sim 110 \text{ cm}^2$ (10.5 x 10.5 cm) and the applied weights ranged from $\sim 150 \text{ g}$ to 10.56 kg, providing a contact pressure range from 130 Pa to 9400 Pa. This covers the range of pressures typically expected from resting a forearm on a desk or countertop to sitting on a surface with full body weight. The cloth receiving surface was mounted on the brass holder and the desired weights (up to 2 kg) were placed onto the top of the wooden block. Together the receiving surface, holder and weights were gently placed onto the contaminated surface such that the entire contaminated surface was covered by the flat area of the receiving surface. The same procedure was used for plastic laminate receiving surface except that weights were placed directly on top of the plastic laminate before it was lowered onto the contaminated surface. Great care was

taken to place the receiving surface down flat onto the contaminated surface without adding any extra force or sliding one surface across the other. After the specified length of contact time, the receiving surface was carefully lifted from the contaminated surface, again being careful to lift it straight up and not to slide one across the other.

For weights over 2 kg, the transfer process was slightly different. The receiving surface was first placed onto the contaminated surface and within a maximum of 2 seconds, the desired weight was placed on top of the wooden block. The weight was removed from the wooden block after the specified contact time and the receiving surface lifted from the contaminated surface ~ 2 seconds later. Thus the two surfaces were in contact for up to a maximum of 4 seconds longer than when the applied weights were less than 2 kg.

2.4 Fluorescein Analysis

The surfaces were placed in 25 x 40 x 4 cm rectangular glass Pyrex dishes into which a 0.1 M buffer solution of Dibasic Sodium Phosphate Heptahydrate ($\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$) was added. The concentration of the buffer solution is important as fluorescein has a pKa (acid dissociation constant) of 6.4 and its ionization equilibrium leads to pH-dependent absorption and emission fluorescent yield over the pH range of 5 to 9. The quantity of buffer solution varied from 100 ml to 400 ml depending on the surface being analysed. The buffer solution was first spread over the surface by tilting the dish backwards and forwards. Next the solution was squirted onto the surface 100 times using a disposable pipette. Seven mL of this washing solution was placed into a cuvette which was then inserted into a Turner model TD-700 fluorometer used to measure the concentration of fluorescein in the solution. The fluorescein concentrations are used to calculate the mass of fluorescein deposited on or transferred to the surface. The fluorometer was calibrated to detect 1 – 125 ng ml^{-1} of fluorescein in the buffer solution. The fluorescence yield (efficiency of the fluorescence process i.e. ratio of number of emitted to absorbed photons) of the fluorescein molecule is very high, excitation occurs at 494 nm and emission at 521 nm.

3 Results and Discussion

3.1 Spatial Uniformity of Deposition in Chamber

Although the fluorescein dispersion system and mixing fan were designed to provide uniform aerosol mixing within the deposition chamber, we also measured the spatial variability in deposited mass to ensure that the surfaces were uniformly contaminated. The spatial deposition within the chamber was measured for each deposition run using the brass plates and found to be uniform to within 4 %.

For all but one deposition run, the ratio of the standard deviation to the average deposited mass on the six brass sheets ranged from 3.3 to 9.5 %. For one run this ratio was 18 %. The average ratio across all the runs was 9.4 %. No systematic differences in the spatial deposition patterns were seen.

3.2 Washing Technique Efficiency

All samples were analysed using the same washing technique as described above (Section 2.4) therefore all results are relative, however it is necessary to quantify the efficiency of the washing technique to understand the uncertainties associated with it. Brouwer et al. (1999) assumed a 100 % recovery rate for their washing technique when a repeated extraction revealed no detectable contaminant. Their technique was to place the loaded plates into a polyethylene tank containing 200 ml of extraction liquid. The tank was shaken for 15 minutes and a 2 ml sample was analysed. This same analysis was conducted on the fluorescein contaminated cotton, fleece, brass and glass samples. Repeated extraction yielded no detectable fluorescein but a more comprehensive method of determining the washing technique efficiency was sought.

Various methods were explored but only one gave consistent results which were achieved as follows: 16 cm² pieces of brass were cut and cleaned using ethanol and buffer solution. These were then weighed on a microbalance (Sartorius Model SE-2) which is sensitive to 0.1 µg and can tare up to 2.0 g. They were then placed in the deposition chamber where they were contaminated with polydisperse fluorescein. The small brass squares were then re-weighed and thus the mass of deposited fluorescein was determined. The pieces of brass were then washed using the same technique as the samples for the transfer experiments and a second value for the mass of deposited

fluorescein was obtained. The comparison of these results gives a percentage recovery value. The average percentage recovery rate for brass is $72 \pm 13 \%$.

It became clear that the absolute humidity of the air has a significant effect on the weight of the brass pieces. A 50 % decrease in the relative humidity lead to an average decrease of $4.5 \mu\text{g}$ in the measured weight of the brass pieces. This is significant as the mass of fluorescein deposited on the brass pieces was of the order of $18 \mu\text{g}$. It was therefore necessary to monitor the humidity and ensure that when weighing the brass pieces before and after the deposition, that room conditions were consistent. This effect however was more prominent on the cotton and fleece pieces and consequently no conclusive results could be resolved. Due to the inconclusive results, a recovery efficiency of 100 % was assumed for all experiments.

3.3 Transfer Efficiencies Between Various Surface Pairs

In this section transfer efficiencies – the percentage of fluorescein on the contaminated surface which is transferred to the clean receiving surface - at a single contact time and pressure for all the surface pairs are compared, except for brass, which was not used as a receiving surface. Throughout the discussion in this section, the first surface mentioned is the contaminated surface and the second mentioned is the receiving surface (i.e. the initially clean surface placed into contact with the contaminated surface). For the mass transfer efficiencies presented in table 1, the contact time is 1 minute and the applied pressure is 890 Pa (i.e. a weight of 1 Kg was placed on the receiving surface of 0.011 m^3).

<i>Receiving Surface</i>	<i>Contaminated Surface</i>			
	Cotton	Fleece	Brass	Plastic Laminate
Cotton	3.1 ± 0.3	5.0 ± 0.5	10.6 ± 1.1	3.4 ± 0.4
Fleece	8.4 ± 1.0	8.2 ± 0.9	41.0 ± 3.2	30.4 ± 2.9
Plastic Laminate	7.2 ± 0.8	2.1 ± 0.2	9.6 ± 1.0	3.5 ± 0.4

Table 1. Mass transfer efficiencies (in percent) between various surfaces with a contact time of 1 min and contact pressure of 890 Pa. The uncertainties are based on the propagation of uncertainties in the experimental method

The transfer efficiencies from contaminated soft surfaces - namely cotton and fleece - to all receiving surfaces will be discussed first. The data in table 1 above show an increase in the transfer efficiency as the receiving surface texture increases in roughness – with the exception of transfer from cotton to plastic laminate. The transfer efficiency from fleece to fleece and from cotton to fleece are similar at ~ 8 %. Transfer efficiencies for fleece to cotton and cotton to cotton are lower. Conversely transfer from fleece to plastic laminate is only ~ 2 % in comparison to over 7 % from cotton to plastic laminate. The transfer efficiencies from the contaminated hard surfaces – brass and plastic laminate - to all receiving surfaces will now be discussed. As shown in table 1, the transfer efficiencies to all three receiving surfaces are higher when the contaminated surface is brass. The highest transfer efficiency observed in our experiments was 41 % for contaminant transfer from brass to fleece. A similarly large value, 30 %, was observed for contaminant transfer from plastic laminate to fleece. In comparison, only between ~ 4 – 11 % of the contaminant is transferred to either plastic laminate or cotton from these hard surfaces, which is slightly more than from soft surfaces.

It is clear that fleece picks up more of the contaminant than either cotton or plastic laminate. At the same time, there is a lower transfer efficiency from fleece to the plastic laminate than there is to cotton. Both phenomena could be due to the surface texture of the fleece. Fleece has a higher fibre pile than does cotton, which could assist in picking up more particles. Also as the fibres of the fleece come in contact with the cotton, there is compression of the fleece fibre pile and thus potentially sideways rubbing of the fleece fibres over the cotton which might allow for the transfer of more contaminant particles. At the same time, the higher fibre pile of the fleece allows for greater penetration of deposited aerosol into the material, which would potentially reduce the number of particles available to be transferred to the hard plastic laminate surface.

3.4 Transfer Efficiency with Increased Pressure

In this section as in the previous, the first named surface is the contaminated surface and the second named is the receiving surface. The contact time is always 1 minute but the applied pressure varies. The graphs show the mean value and the error bars indicate the propagation of uncertainties in the experimental method.

3.4.1 Cotton to Cotton

Figure 2 presents the transfer efficiency from contaminated cotton to clean cotton with increasing pressures. Based on the data in figure 1 – and as suggested by data in the subsequent figures – there appears to be two mass transfer efficiency regimes – a low efficiency of ~ 2 – 4 % and a high of ~ 6 – 10 %. The transfer efficiencies within each regime are relatively independent of pressure in their respective pressure range, with a transition pressure region between the two pressure ranges. For the cotton-to-cotton surface pair, this transition pressure is ~ 3500 Pa. Detailed experiments in the vicinity of this pressure could reveal how sharp this transition is.

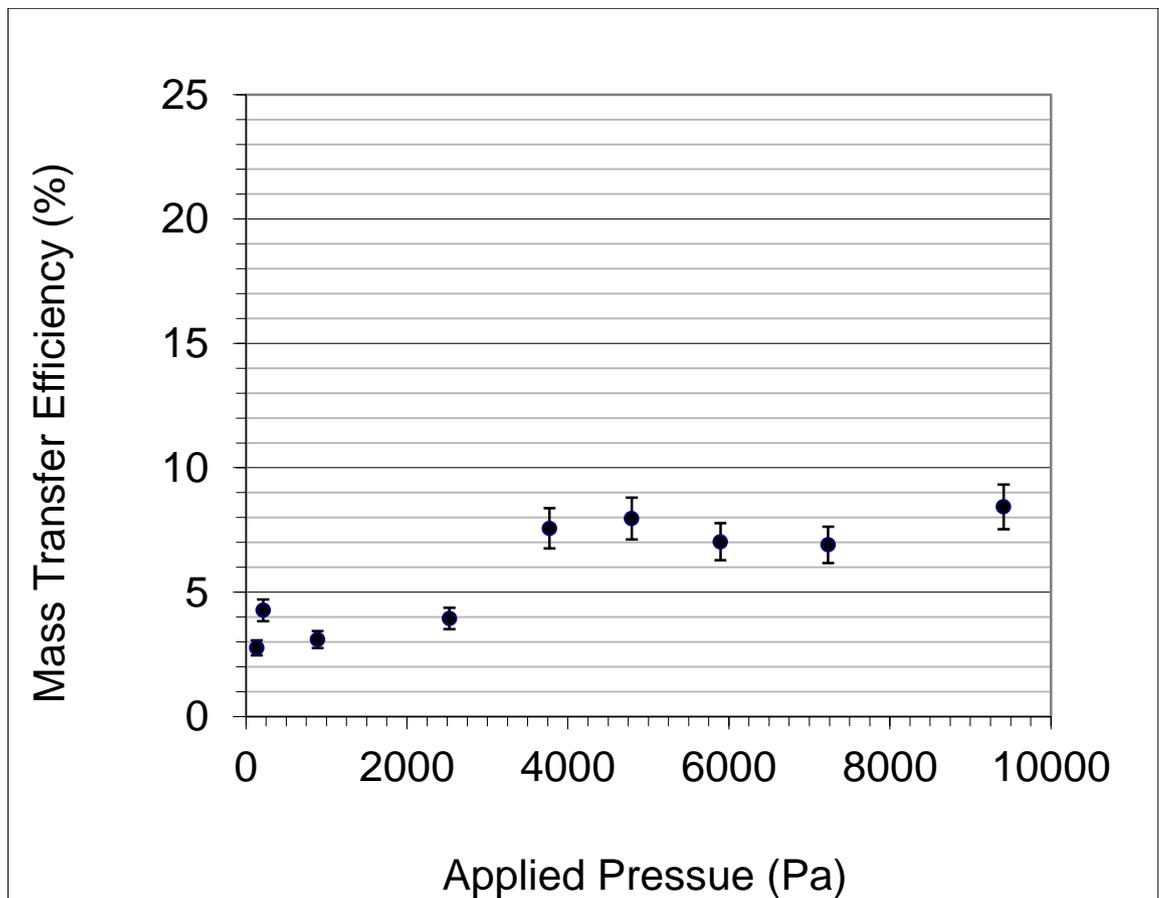


Figure 2. Cotton to cotton mass transfer efficiencies as a function of applied pressure for 1 minute contact time

3.4.2 *Fleece to Fleece*

Mass transfer efficiencies as a function of applied pressure for contact between contaminated fleece and clean fleece are illustrated in figure 3 and show a pattern similar to that seen for the cotton to cotton contact transfer. As with cotton, the transfer efficiencies are essentially independent of applied pressure in the two different pressure regimes. The low and high pressure transfer efficiencies are more than a factor of two higher, $\sim 7 - 9$ Pa and $\sim 15 - 18$ Pa, respectively than those measured for cotton-to-cotton and the transition pressure is lower, ~ 1700 Pa.

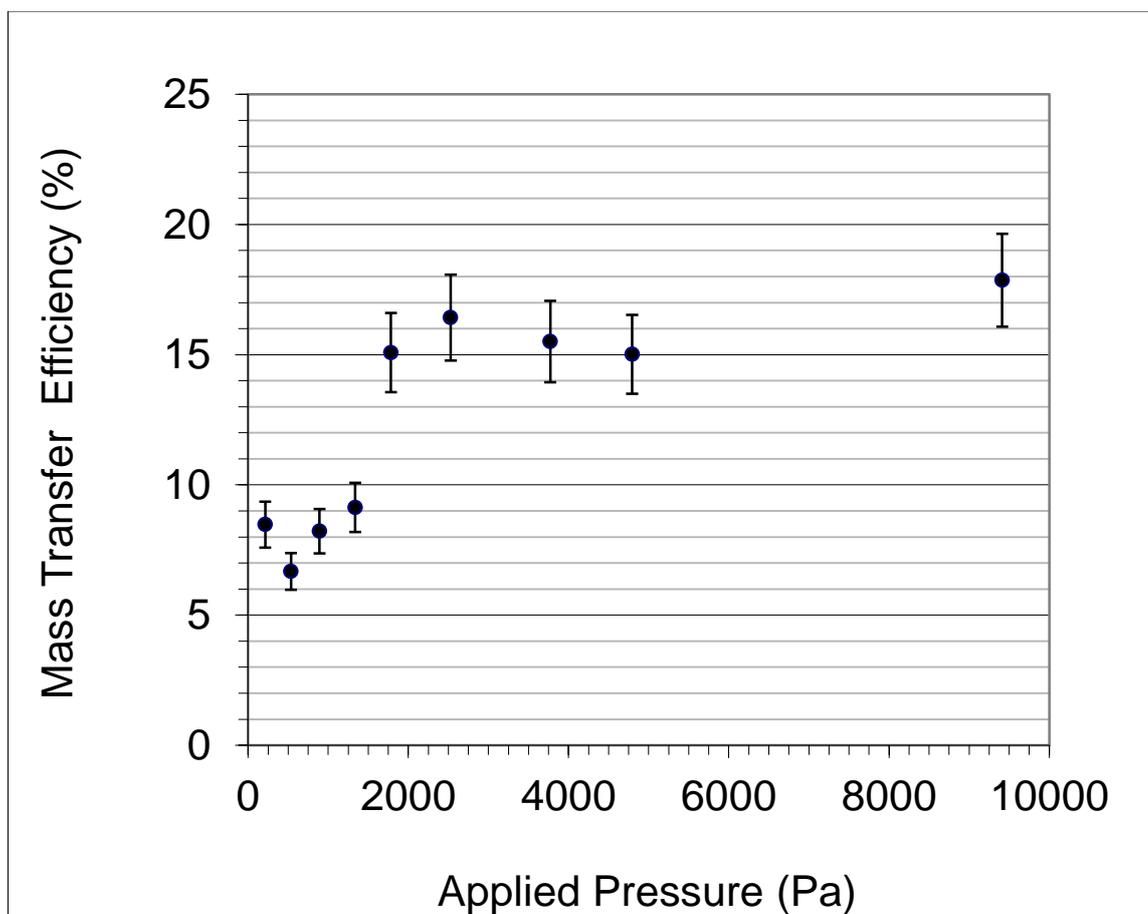


Figure 3. Fleece to fleece mass transfer efficiency as a function of applied pressure for 1 minute contact time

3.4.3 Cotton to Fleece

Identical experiments to those described above were carried out with cotton as the contaminated surface and fleece as the receiving surface. The resulting data are shown in figure 4. The transfer efficiency exhibits a pattern similar to that observed above. The transfer efficiencies at the lower pressures are ~ 5 % and ~ 8 to 11 % at the higher pressures; in this case, the transition pressure is ~ 600 Pa, the lowest inferred in these experiments.

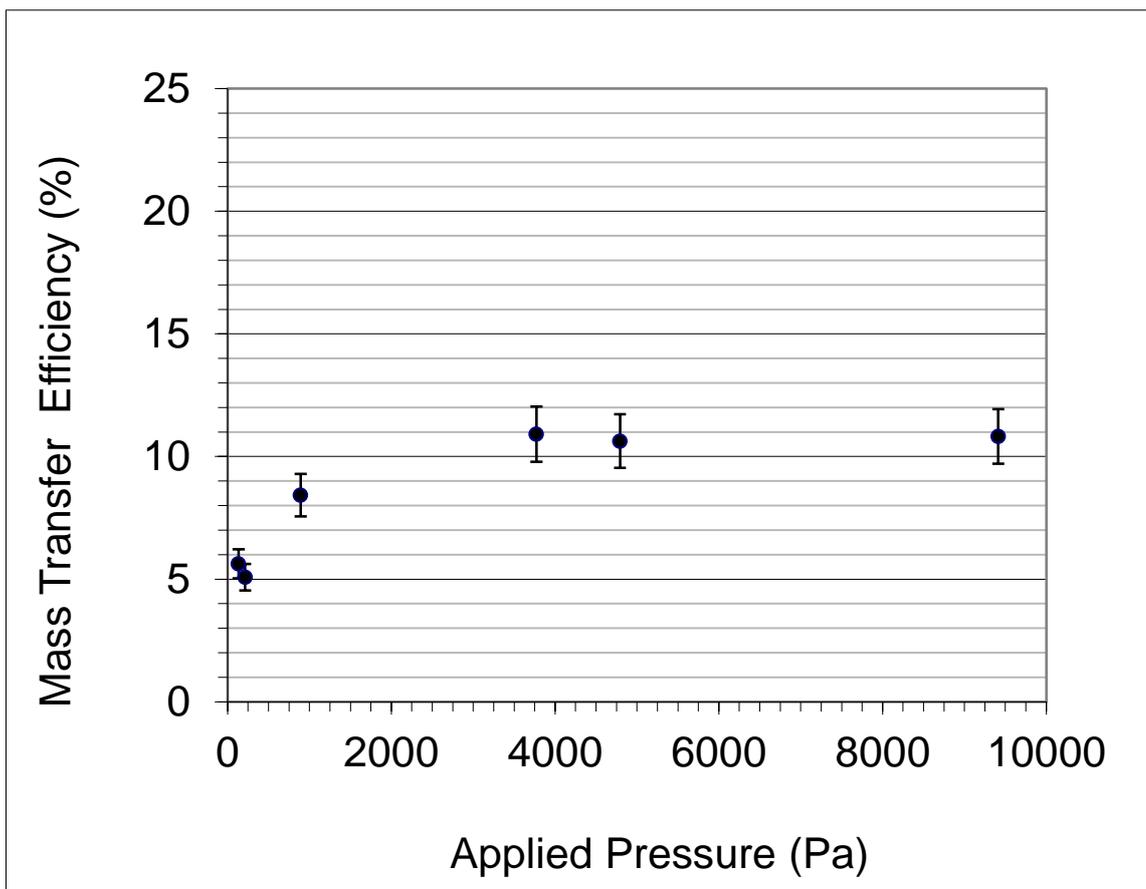


Figure 4. Cotton to fleece mass transfer efficiency as a function of applied pressure for 1 minute contact time

3.4.4 *Plastic laminate to Fleece*

Figure 5 is a graph of the percentage transfer when contaminated plastic laminate comes into contact with clean fleece, for a contact time of 1 minute and for various pressures. For all but the transfer efficiency at the highest applied pressure, the transfer efficiency vs. applied pressure pattern is similar to that observed for the other surface pairs – a low pressure and a high pressure regime where the mass transfer efficiencies are relatively constant $\sim 9 - 13 \%$ and $\sim 24 - 31 \%$ respectively. The transition pressure is ~ 900 Pa. The highest transfer efficiency – observed at the highest applied pressure of 9400 Pa – is 44% , which is higher than the efficiencies observed above the 900 Pa transition pressure. This highest efficiency is the average of 2 measurements, so cannot be easily attributed to measurement error, etc. It is possible that there is another pressure regime defined for pressures above ~ 9000 Pa, which was not explored in these experiments.

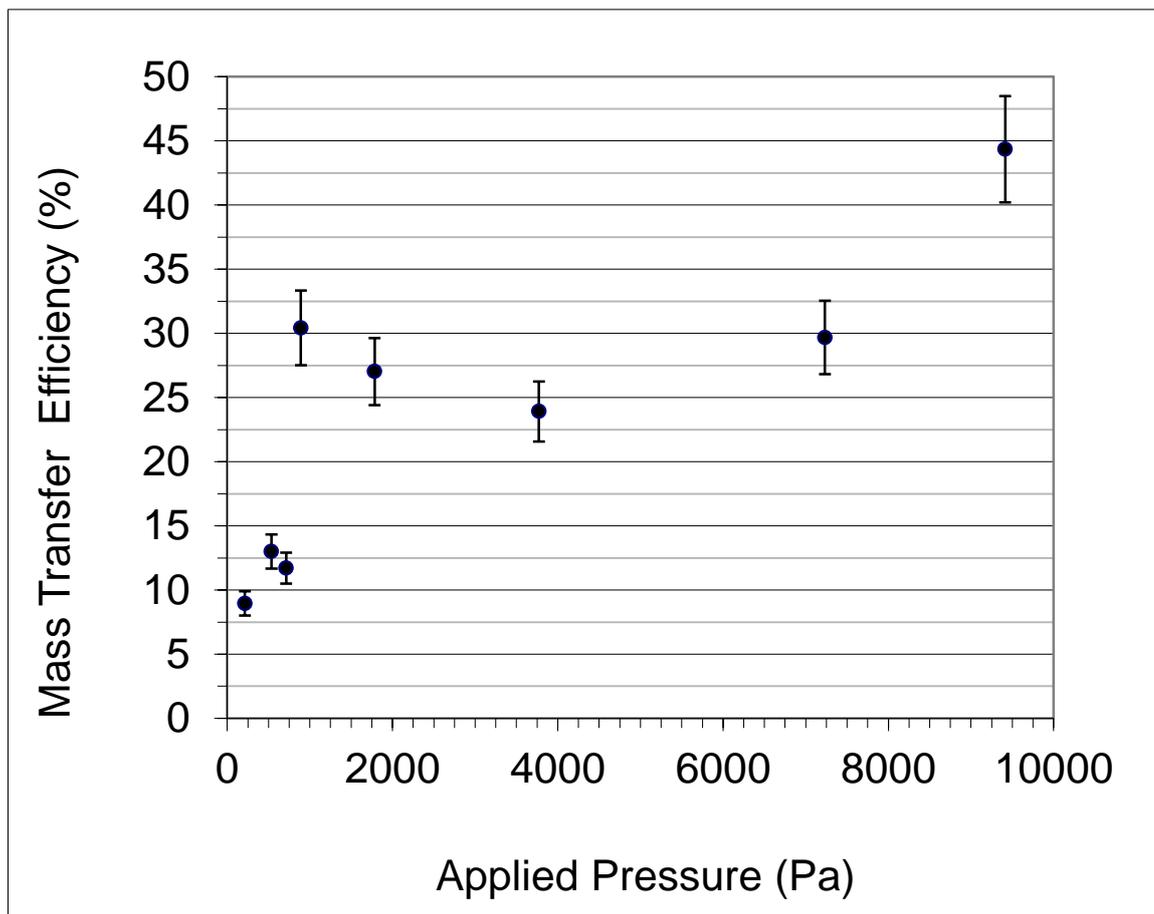


Figure 5. Plastic laminate to fleece mass transfer efficiency as a function of applied pressure for 1 minute contact time

3.5 Transfer Efficiency with Increased Time

We investigated whether the mass transfer efficiency changes as a function of contact time. In these experiments, we examined only the cotton-to-cotton surface pair at two applied pressures, 890 and 4500 Pa. These two pressures are in the two pressure regimes we discussed earlier. The results are shown in figure 6, where the time scale is logarithmic to accommodate the large range of contact times from ~ 2 seconds to 1 hour. There appears to be no clear trend in the transfer rate between contaminated cotton and clean cotton with increased time at either of the two pressures. There could be a slight increase in transfer efficiency with time with time for the data collected at an applied pressure of 4500 Pa, where the transfer efficiencies observed for contact times of 1 min or less are ~ 7 to 8 % compared with the ~ 10 % efficiency at contact time of 1 hour. However, as can be seen in the figure, the transfer efficiencies for the same applied pressure of 4500 Pa, drop to between 5 and 6 % for contact times of 5 and 30 minutes. Again, these data are averages of 2 - 4 measurements, so random measurement error does not appear to provide an explanation.

As an interesting side note, these results shed some additional light on the ‘5 second rule’ (“if food falls on the ground, it may be safely eaten as long as it is picked up within five seconds” – Wikipedia, accessed Nov 2011). The results here indicate that providing there are no external forces, the length of contact time between a clean and contaminated piece of cotton has essentially no impact on the percentage of contaminant transfer, even at very short contact times. These results are consistent with experiments done by Dawson et al. (2007), who examined the ‘five second rule’ using pieces of food and surfaces inoculated with bacteria.

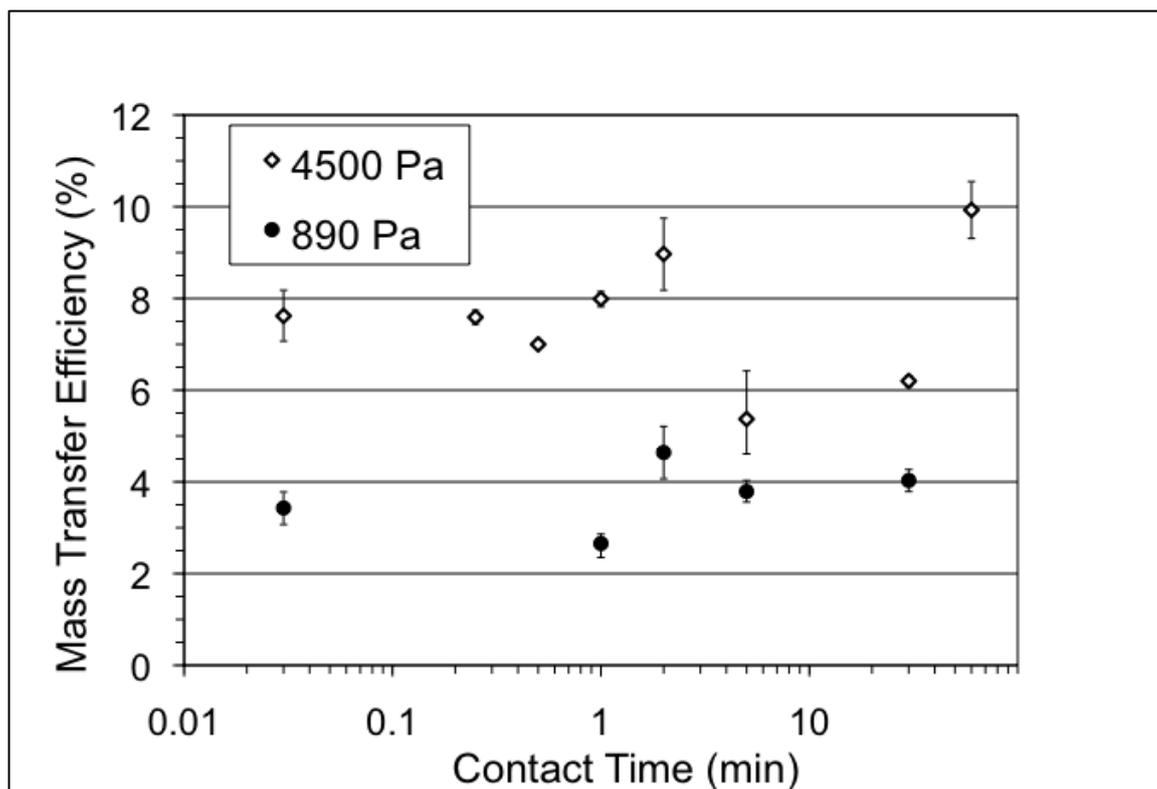


Figure 6. Cotton to cotton mass transfer efficiency as a function of time for two constant applied pressures, 4500 and 890 Pa. Error bars show the range of the observed transfer efficiency values

3.6 Transfer Efficiency with Increased Contaminant Loading

We examined the effect of change in contaminant loading on mass transfer efficiency for several different values of applied pressure. These experiments were conducted for only one surface pair, cotton to cotton, and for a constant contact time of 1 min. For the ‘low mass loading’, fluorescein was deposited onto cotton surfaces in the deposition chamber using the procedure described in section 2.2, yielding an average deposited mass of 1560 ng cm^{-2} . For the ‘high mass loading’ case, fluorescein was injected into the chamber until the APS indicated an airborne concentration of $1200 \text{ particles per cm}^3$ – the single particle detection limit. The airborne particles were then allowed to deposit for approximately 35 minutes until the airborne particle concentration was $\sim 300 \text{ particles per cm}^3$, at which time the generator was turned on again. This process was repeated 5 times. The average particle loading on the contaminated cloth (determined

from the washing technique) for the high loading case was 5740 ng cm^{-2} , approximately 3.7 times higher than the low mass loading case.

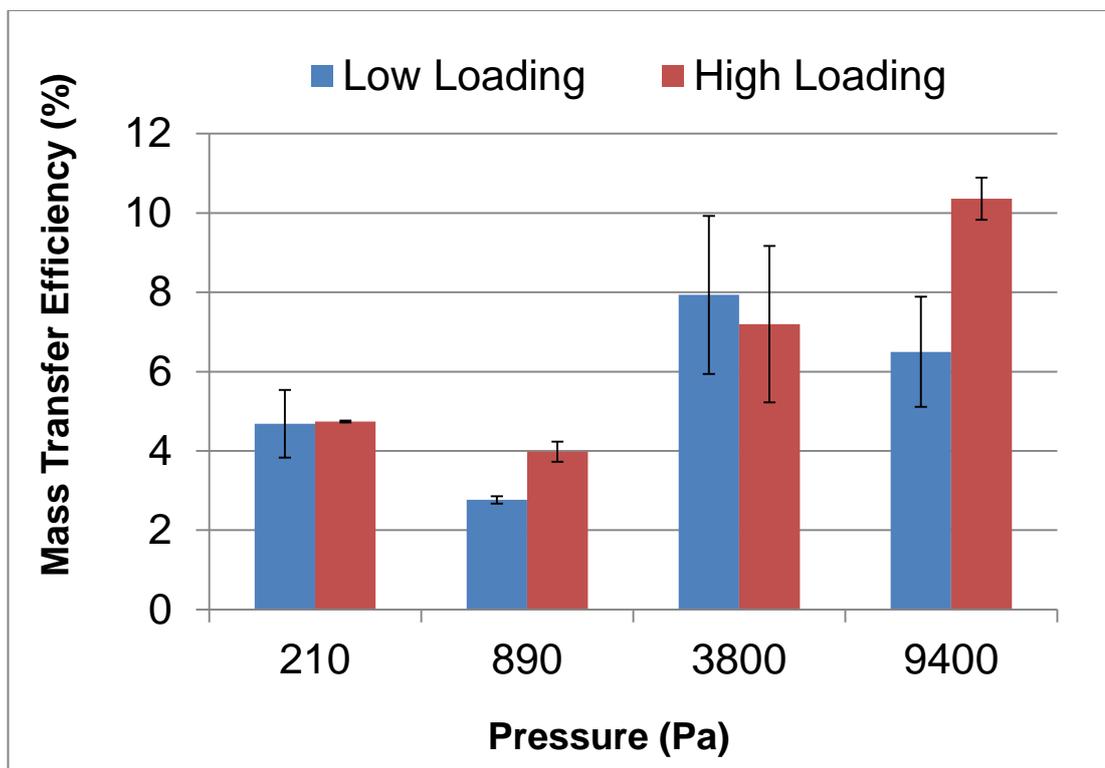


Figure 7. Cotton to Cotton transfer efficiency for a contact time of 1 min at a given applied pressure, for low or high particle loading. The graph indicates the mean and the range of transfer efficiency values

Based on the data shown in figure 7 there appears to be a slight difference in the mass transfer efficiency for low versus high contaminant loading at two of the four applied pressures, and essentially no difference at the other two pressures. One might have expected that with a significant increase in the mass of contaminant deposited on a surface proportionally more mass would be available for transfer, especially at higher contact pressure. However, there appears to be no discernable trend or systematics. Additional experiments are necessary to show whether other surface pairs behave similarly and if a larger range in mass loading may yield different results.

4 Summary and Conclusions

Experiments were carried out to quantify the fraction of mass transferred from a particle-contaminated surface to a clean surface. A matrix of four variables was investigated: different surface pairs, applied pressure, contact time and contaminant loading. Among these, the variable with the greatest influence on mass transfer efficiency was the choice of surface pairs. Two types of surfaces were investigated, two soft surfaces – cotton and fleece – and two hard surfaces – brass and plastic laminate. The lowest mass transfer efficiency was observed for transfer from contaminated fleece to plastic laminate, ~ 2 %, and the highest efficiencies were seen for transfers from the two hard surfaces, brass and plastic laminate, to fleece, 41 and 30 % respectively. While these initial experiments were not designed to investigate the details of the transfer process, the data suggest that the surface texture of the fleece both enhances the transfer of particles from the other surfaces to the clean fleece and inhibits particle transfer from contaminated fleece.

This concept of particles being easier to remove from a hard surface versus a soft surface has been observed by in the related area of particle resuspension from surfaces. Although contact transfer and resuspension aren't the same mechanistically, they both share a similar concept – that of having particles 'available' for transport. Braun et al. (2002) looked at the resuspension of particles from soft versus hard flooring surfaces and found that soft surfaces (carpeting) can act as a filter by trapping and holding the particles thus making them less available for resuspension in comparison to a hard flooring surface. Similarly, Andersson et al. (2004) observed greater resuspension from wood and plastic than from carpeting. Wille (1974) established that it requires approximately ten times the air velocity to resuspend a settled particle from a carpeted surface versus a hard surface.

Correspondingly, soft surfaces tend to accumulate or 'pick up' more particles than a hard surface i.e. there appears to be increased transfer with increased surface texture or roughness. This is consistent with the observations of Braun et al. (2002) and Wille (1974) who concluded that once a particle encounters a three-dimensional soft fabric it becomes mechanically trapped and therefore must be dislodged if it is to be resuspended. When investigating the transfer efficiency between various surfaces as a function of contact pressure, it became apparent that the data could be described as having two pressure regimes (as opposed to a consistent increase in transfer efficiency with

increasing pressure), one at low pressure and one at high pressure where the mass transfer efficiencies are relatively constant as a function of applied pressure within each regime. A transition pressure region connects the two pressure regimes. The transfer percentages for the various surface pairs and the transition pressures are summarised in table 2.

Surface		Average Transfer Efficiency (%)		Transition Pressure (Pa)
Contaminated	Receiving	Low P	High P	
Cotton	Cotton	3	8	3500
Fleece	Fleece	8	16	1700
Cotton	Fleece	6	11	600
Plastic laminate	Fleece	12	30	900

Table 2. Summary of transfer efficiency results as a function of applied pressure

To put these transition pressures into context, the lowest transition pressure of 600 Pa is equivalent to an applied mass of ~ 700 g and the highest of 3500 Pa is equivalent to an applied mass of ~ 4 kg. A forearm resting on a desk would yield approximately 2675 Pa of pressure (~ 3 kg) and the average adult sitting on a chair would exert a pressure of ~ 44,490 Pa (~ 50 kg). Thus simply resting a person's arm on a desk would provide enough pressure to move into the higher transfer efficiency regime.

The length of time the clean and contaminated surfaces are in contact appears to have no effect on the fraction of contaminant transferred between them. This is providing there are no external forces e.g. 'rubbing' of the two materials etc. The contact time ranged considerably from ~ 2 seconds to 1 hour. This observation agrees with Dawson et al. (2007) but disagrees with Vatistas (1992) who concluded that the adhesion of particles to surfaces increases with time, due to the gradual increase in the contact area over which the attractive forces are operative.

The mass of deposited particles on the contaminated surface does not appear to significantly influence the mass transfer efficiency – at least for the cotton to cotton surface pair we examined here. This disagrees with the findings of Brouwer et al. (1999) who concluded that the level of contaminant transferred to the skin is dependent on the level of surface contamination. However Brouwer’s mean low and high loadings were $6 \mu\text{g cm}^{-2}$ and $177 \mu\text{g cm}^{-2}$ respectively, while our mean low and high loadings were $1.5 \mu\text{g cm}^{-2}$ and $5.7 \mu\text{g cm}^{-2}$. Therefore the actual particle densities for the loadings used in this experiment are still quite low and thus the nature of the surface layer of the particles unlikely to have changed. This could account for the inconclusive change in transfer with change in surface loading reported here. Brouwer et al. (1999) also reported transfer efficiencies from glass to skin of $\leq 2 \%$ which are less than the lowest transfer efficiency recorded in this study.

Overall, the observed mass transfer efficiencies of 2 – 45 % are very significant numbers in terms of aerosol mass transport in the environment. If an indoor environment is contaminated – especially if that contamination goes unnoticed – the contamination could be spread in large quantities via the process of contact transfer thus increasing a persons’ exposure risk. These experiments were designed as an initial quantitative exploration using a limited number of surfaces and study variables. Two variables not studied here that may be important are the effects of surface charge and different aerosols shapes, especially biological aerosols. These results also suggest that additional studies should be conducted to better understand the details of the transfer process or processes.

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General Conclusion

1 Summary of Results

The purpose of this work was to experimentally generate data to quantify the levels of hazardous airborne particles associated with secondary exposure. Two transport processes were examined: the resuspension of particles from clothing surfaces and the contact transfer between various indoor surfaces of formerly airborne hazardous particles.

This section discusses the key findings from the studies as detailed in the preceding sections.

1.1 Resuspension

A study was conducted with the aim of quantifying the resuspension of supermicron sized particles from various clothing surfaces. The variables investigated include particle size, contaminated clothing type, body location of contaminated clothing, level of physical activity engaged in by contaminated person, and variation in resuspension rates with time. The influence of each of these variables on the levels of particle resuspension, was deliberated upon in the previous sections, and a brief discussion of the key results will be discussed below.

1.1.1 Increasing particle resuspension with increasing particle size

From theory, it is expected that the likelihood of particles resuspending from a surface will increase with increasing particle size. However to date, there have been no experimental studies conducted to investigate this theory in relation to resuspension from clothing surfaces due to human physical activity. The results obtained in this study confirm that a higher quantity of larger sized particles will resuspend from clothing surfaces, than will smaller sized particles, as a result of human physical activity. This has been verified in three ways:

- 1) Comparison of the size distribution curves of airborne particles during deposition and during resuspension, disclosed a marked difference in their shapes. The highest mass concentration of airborne particles during deposition occurs for the 3 μm sized particles and the smallest at 10 μm , while the highest

mass concentration during resuspension occurs for the 10 μm sized particles and the smallest at 3 μm . This indicates that of the particles deposited onto the clothing surface (which are therefore available for resuspension), more of the larger sized particles resuspended than did the smaller particles.

- 2) Examining the individual size distributions for each of the three particle sizes investigated revealed a shift towards a higher Mass Median Aerodynamic Diameter (MMAD), for the resuspended distributions, in comparison to the deposited distributions. For the 3 μm distribution, a 0.12 % increase in the diameter of resuspended particles in comparison to those deposited was observed. The 5 μm distribution displayed a 0.16 % increase and the 10 μm distribution revealed a 0.09 % increase in the resuspended particles' median diameter in comparison to that of the deposited particles.
- 3) The fraction of deposited particles which resuspended from clothing during human physical activity, as determined via Neutron Activation Analysis (NAA), increased with increasing particle size. The resuspended fraction (RF) varied depending on the physical activity level. During low physical activity, an average of 27 ± 7 % of 3 μm particles, 28 ± 7 % of 5 μm particles and 30 ± 8 % of 10 μm particles resuspended. During high physical activity, an average of 27 ± 9 % of 3 μm particles, 30 ± 6 % of 5 μm particles and 34 ± 5 % of 10 μm particles resuspended.

Although the results from Aerodynamic Particle Sizing (APS) and Neutron Activation Analysis (NAA) both show an increase in resuspension with increasing particle size, only the results from the APS show a statistically significant increase (at $p = 0.05$). This apparent discrepancy is due to the nature of the analysis techniques and their capturing of different features of the resuspension process, as discussed in previously. The notable difference is that the samples analysed by NAA include particles that have arrived at a clothing surface via inertial impaction, and this feature is not detected by the APS.

1.1.2 Relationship between particle size and the height reached by resuspended particles

If a person wearing contaminated clothing engages in any form of physical activity, it will result in particles resuspending from the clothing surface. This will change a person's potential exposure route from contact exposure (through the skin) to inhalation. Therefore the height reached by the resuspending particles, and the size distribution of the particles at that height, is of utmost importance in determining the potential severity of a person's exposure. The mass concentration of resuspended 3, 5 and 10 μm sized particles were examined at three heights: ankle (15 cm), waist (90 cm) and head (150 cm) heights, and for the 3 μm sized particles, their highest concentration was found to be at ankle height. The opposite is true for the 5 and 10 μm sized particles, whose concentration increased as height above the ground increased. This finding, of high concentrations of larger sized particles being present at head height, has consequences for a person's potential inhalation exposure, as discussed in previously. As larger particles are less likely to penetrate the lower airways of the lungs, they therefore do not pose a major threat for increased inhalation exposure. Furthermore, as the smaller sized resuspended particles, which are more hazardous in terms of inhalation exposure, were found in higher concentrations at ankle height, they are likely to deposit to the ground and thus could be more easily removed from the environment by cleaning etc.

1.1.3 Airborne mass concentration variation with time, during resuspension

An APS was used to determine the concentration and size distribution of seeded particles which were subsequently resuspended from the clothing of a physically active person. The APS was set to measure the airborne particle concentration every second. A real-time rate of change of airborne particle concentration (represented in $\text{mg m}^{-3} \text{min}^{-1}$) can therefore be determined.

The data was found to be separable into two separate regimes: the first having a high, positive rate of change of airborne particle concentration relative to the second regime, and occurring within the first 1.5 minutes of the beginning of the resuspension event. The second regime revealed a slower rate of change of particle concentration and remained relatively unchanged for the remainder of each resuspension event.

For the first regime, both physical activity levels result in an increase in airborne particle concentration, but high physical activity results in a rate of increase four times that corresponding to low physical activity. For the second regime, the change in airborne particle concentration shows an opposing trend for low compared to high physical activity levels. It can therefore be concluded that following deposition of hazardous aerosol particles to a person's clothing, one is at highest risk of secondary exposure, during the first 1.5 minutes of physical activity. The exposure risk after this time is dependent upon the level of physical activity engaged in by person.

1.1.4 Resuspension with varied physical activity level

Unless a person remains perfectly still, they will engage in some form of physical activity. The level of activity is highly variable. This study aimed to quantify the influence of various physical activity levels on the resuspension of particles from a person's clothing. Two physical activity levels were investigated: low physical activity which consisted of walking at a pre-defined rate for 20 minutes, and high physical activity which was representative of running and simulated by modified Irish dancing to a Reel for 10 minutes, as previously described.

Following NAA analysis, the RF of particles at low physical activity (averaged over all particle sizes and contaminated material types) was found to have an average value of 28 ± 8 %. The range of RF values during high physical activity was found have an average value of 30 ± 7 %. Statistical analysis of the average RF values at low and high physical activity levels revealed no significant difference. However, examining APS data for the same parameters revealed a tenfold increase in the cumulative mass of airborne particles during high physical activity in comparison to that during low physical activity, and this difference was statistically significant.

1.1.5 Resuspension from specific body locations

The contaminated clothing surfaces were attached to the volunteer at four different body locations, to examine if the location on a person's body which is contaminated will influence the level of particle resuspension.

Statistical analysis determined that there was no significant difference in the RF between samples obtained from different body locations, at low physical activity.

However at high physical activity, there was a statistically significant difference in the RF between samples obtained from different body locations, and the largest and most significant difference existed between the RF at the front and the back of the legs.

1.1.6 Resuspension with varied contaminated clothing material type

The resuspension of particles from four different clothing material types was examined, to determine if specific fibre types (natural or synthetic), or material weave patterns, would aid or hinder particle resuspension from their surface. The material types used included cotton, polyester (a synthetic fibre but with the same weave pattern as cotton), fleece (polyester but with a very different weave pattern), and denim (a textile with a mixture of natural and synthetic fibres). The results revealed that the fibre type had no influence on the levels of particle resuspension from their surface, but the material's weave pattern (and hence the material's surface texture) did significantly influence the levels of particle resuspension. The level of resuspension of formerly deposited particles between material types was significantly different from fleece, and fleece alone. It was determined via Scanning Electron Microscopy (SEM) that cotton, polyester and denim were all weaved in the same pattern and hence have a comparable surface texture, while fleece was found to have a very different weave, with a non-uniform pattern, and hence had a much rougher surface texture.

1.2 Contact Transfer

A study was conducted with the aim of quantifying the mass transfer efficiency (TE) of deposited aerosol particles (diameter range 0.5 – 20 μm , with a MMD of $12.9 \pm 3.8 \mu\text{m}$) when selected soft and hard surfaces come in contact. The variables investigated include surface pairs in contact, applied pressure, contact time, and contaminant loading. The influence of each of these variables on the mass transfer efficiency (TE) was deliberated upon previously, and a brief discussion of the key results will be discussed below.

1.2.1 Transfer efficiencies between various surface pairs

Four different surface types were used to investigate the effect which the contaminated and receiving surface types had on the TE of particles between the surfaces. Two soft surfaces were used: cotton and fleece, and two hard surfaces: plastic laminate and brass.

In this section, the first mentioned surface is the contaminated surface and the second mentioned surface is the initially ‘clean’ receiving surface. Contact time and pressure were kept constant.

TE’s to all surface types, from contaminated soft surfaces, ranged from 2.1 to 8.4 %, while TE’s from contaminated hard surfaces ranged from 3.4 to 41.0 %. Following analysis, a general trend emerged, revealing that there was an increase in the TE as the *receiving* surface texture increased in roughness.

The lowest TE was observed for transfer from contaminated fleece to plastic laminate, 2.1 ± 0.2 %, and the highest TE’s were seen for transfers from the two hard surfaces, brass and plastic laminate, to fleece, 41.0 ± 3.2 % and 30.4 ± 2.9 % respectively. The data indicated that ‘clean’ fleece picked up more contamination from a surface and also that ‘contaminated’ fleece held the particles more tightly than the other surfaces. Both these findings were attributed to the comparatively rough surface texture of the fleece material.

1.2.2 Transfer efficiency with increased pressure

The TE between various surfaces as a function of contact pressure was investigated. The applied contact pressure ranged from 130 Pa to 9400 Pa. The data revealed two pressure regimes (as opposed to a consistent increase in TE with increasing pressure), one at low pressure and one at high pressure, where the TE’s were relatively constant as a function of applied pressure within each regime. A transition pressure region connects the two pressure regimes. The TE’s in each pressure regime and the transition pressures varied depending on the surface pairs in contact. The low pressure regime TE’s ranged from 3 – 12 % and the high pressure TE’s ranged from 8 – 30 %. The transition pressures ranged from 600 to 3500 Pa, which are low pressures relative to human behavior indoors, the higher pressure being comparable to resting a forearm on a desk.

1.2.3 Transfer efficiency with increased time

The influence on TE of the length of time which clean and contaminated surfaces are in contact was investigated. The contact time ranged considerably from ~ 2 seconds to 1 hour. The results indicated that the contact time of a clean and contaminated surface, had no effect on the fraction of contaminant transferred between them, even at very

short contact times. This was providing there were no external forces e.g. ‘rubbing’ of the two materials etc.

1.2.4 *Transfer efficiency with increased contaminant loading*

The effect of change in contaminant loading on TE for several different values of applied pressure was investigated. The experiments were conducted for only one surface pair, cotton to cotton, and for a constant contact time of 1 minute. For the ‘low mass loading’, the average particle loading on the contaminated cloth (determined from the washing technique) was 1560 ng cm^{-2} . For the ‘high mass loading’, the average particle loading was 5740 ng cm^{-2} . This was approximately a 3.7 fold difference in contamination levels between low and high mass loading cases. For the surface pairs and contact pressures examined in this study, there appears to be no discernable trend in TE variation as a function of contaminant loading.

2 General Discussion

The data presented in this thesis has shown that, when a person wearing contaminated clothing engages in even low levels of physical activity (simply walking), up to 52 % of that contamination can be resuspended from their clothing. The resuspended contamination is then suspended in the air surrounding the person, thus increasing their potential for inhalation and dermal exposure. The most pronounced contribution of resuspension to exposure risk was observed under the following conditions: during the first 1.5 minutes of a person wearing contaminated clothing of fleece material, and while engaging in a high level of physical activity.

In terms of inhalation exposure risk from resuspended aerosols, it was revealed that, at a distance of 75 cm from the source and at head height, a fourfold increase in concentration of 5 and 10 μm sized particles was observed, in comparison to 3 μm sized particles. This indicates that resuspended material is of lesser consequence for inhalation exposure, as the larger particles which are seen are higher concentrations at head height, are less inhalable. This is compounded by the results in “*A Study of the Size Distribution of Aerosol Particles Resuspended from Clothing Surfaces*”, which validate the theory that larger particles are more likely to resuspend than are smaller particles.

The conceptual model of Thomas Scheider and colleagues for assessment of dermal exposure (Schneider et al., 1999), clearly states the importance of having a model properly parameterized, in order to accurately predict exposure. On publication of the Schneider et al. (1999) paper, it was acknowledged that measurement of mass, concentration, and the transport processes had to be based on a theoretical model, as methods for measuring uptake were not available. This study has provided a method for measuring particle uptake from a contaminated surface to a clean surface. Schneider's model was categorized into six compartments and two barriers, together with eight mass transport processes. The present research has provided further information regarding the 'source' compartment, by quantifying the mass of contaminant introduced into the other compartments (e.g. to the air) for specific levels of physical activity. Furthermore, the data presented in this thesis has added to the knowledge base of parameter values for two transport processes; resuspension and contact transfer; by quantifying the potential significance of each of these transport processes. Specifically, this work has provided quantification of the potential contribution of one of Schneider's resuspension definitions: 'the mass of hazardous substance lost from the outer clothing contaminant layer of a particular worker to the air compartment by resuspension per event', and one of the transfer definitions: 'the mass of hazardous substance transferred from the surface contaminant layer to the outer clothing contaminant layer for a particular worker by direct contact per event'.

The present study has provided some insight into the potential for a contaminated person to act as a 'fomite' (Burkhart and Burkhart, 2007, Williams, 2009) by placing a second person (in close proximity to the fomite) at risk of exposure due to the resuspension of particles from the fomite, to the air surrounding the second person. The results of the variation of airborne mass concentration with time, during resuspension, indicated that following exposure, the highest rate of increase of airborne particle concentration (due to resuspension) occurred within the first 1.5 minutes of activity. Therefore during this time, a second person is at highest risk of exposure from particles resuspending from a contaminated person. Following this initial 'high risk' period, the risk to the second person varies depending on the level of physical activity engaged in by the contaminated person.

In relation to a choice of clothing material type for use in high risk contamination scenarios, it was revealed that fleece material behaves significantly different from other

material types investigated. It was concluded that this was due to the nature of the surface texture roughness of the fleece material. In the resuspension study, fleece was found to allow the highest levels of particle resuspension, as particles did not become trapped deep within the weave pattern of the material and were thus relatively easily released from the weave of the material, when the material was subjected to agitation. The contact transfer study revealed that fleece both collected (for clean fleece in contact with various contaminated surfaces) and retained (for contaminated fleece in contact with various clean surfaces) the highest levels of contamination. Again, this was attributed to the weave pattern of the fleece material, as the non-uniform and erratic nature of the weave resulted in the contamination not being distributed in a monolayer, which did not facilitate easy transfer of surface layer contamination to other more uniformly structured weaved materials. These results combined indicate that people wearing clothing made of a fleece material, who were unexpectedly exposed to hazardous aerosol particles, would be at higher risk for re-distribution that contamination if they engage in physical activity. However, it also indicates that for decontamination purposes, a cloth of fleece material would collect deposited aerosol particles in higher concentrations than other material types, and if not vigorously agitated, would retain the particles for efficient removal from the environment.

In conclusion, this study has quantitatively proven that both Resuspension and Contact Transfer are efficient means of re-distributing contamination in the environment. Results have shown that physical activity can cause up to 67 % of contamination deposited on clothing, to be resuspended back into the air and is then a potential for re-exposure. Furthermore, the observed mass transfer efficiencies of up to 45 % are very significant in terms of aerosol mass transport in the environment.

3 Limitations of the studies

Both the resuspension and contact transfer studies were experimental in nature and while designed to be representative of real life scenarios, there were inevitable limitations, which are outlined below.

This study was limited to examining supermicron sized particles, due to the choice of base particles in both the resuspension and contact transfer investigations. Silica was chosen due to its availability in a number of monodisperse sizes and also due to its porous surface which was necessary in order to attach the rare earth metal tracers. The smallest size available in monodisperse, porous silica was 3 μm . The polydisperse aerosol fluorescein, was the chosen particle for the contact transfer study. This was due to the analysis facilities readily available in the Lawrence Berkeley National Laboratory, where the study was conducted. Fluorimetry was relatively inexpensive and it was possible to analyse an abundance of samples in a very short space of time. While supermicron particles are representative of many types of hazardous particles in the atmosphere, an inclusion of submicron sized particles would have further enhanced the study. Examples of hazardous aerosols of submicron size are volatile radioactive aerosols, some viruses and some endospores.

The Neutron Activation Analysis technique required the placing of samples to be analysed into one centimetre high capsules, the largest size capsule available. This involved the rolling or folding of contaminated clothing samples and 55 mm filter papers, which were then crammed into the capsules. While great care was taken to disturb the samples as little as was possible, this may have added a small degree of error to the calculated levels of contamination on the samples. Any samples which were deemed to have been disturbed in any way were discarded.

The experiments were devised so that the two analysis techniques, APS and NAA, were operational simultaneously, and used to derive comparable results for a number of variables investigated. While both techniques did reveal the same trend in their results for the same parameter, the APS data alone provided statistically significant results. It was concluded that this was due to the analysis techniques identifying different parts of the resuspension process. This was an interesting finding and should be taken into consideration when designing future experiments involving either or both of these analysis techniques.

During the resuspension events, active and passive filters were operational at various heights, to determine the size distribution of particles detected at ankle, waist and head heights. The data, were interesting and unexpected (it was expected that the highest concentration of larger particles would be seen at ankle height, due to the gravitational force having an increasing effect with increasing particle size). One possible reason for the unexpected findings was the short distances (75 and 120 cm) from the physically active contaminated person, at which the filters were placed. These short distances were chosen to ensure a detectable level of resuspended particles would be captured by the filters. However, if the filters were placed at further distances from the source, a different size distribution may have been observed at the various heights, as gravitational settling would have time to be a more dominant factor and hence more of the larger particles may be seen at lower heights from the floor.

For the contact transfer study, while great care was taken to place and remove the surfaces in contact without adding extra pressure or dropping one surface onto the other, or sliding one surface across the other, there was undoubtedly some degree of error associated with this method. For future studies, a mechanical system could perhaps be devised to better control the placing, and removal, of the surfaces in contact.

Examining the transfer efficiency of particles between two surfaces in contact with respect to changes in contact time and contaminant loading revealed no discernable trends. The extent of the study of these variables was limited due to a limited time period in which to carry out the experimentation, but both variables warrant further investigation, with an extension of the range of values examined in each variable and also further replicates of each trial.

4 Future Research

Future research into the resuspension of hazardous aerosol particles from human body surfaces could involve a 'real' contamination scenario. To ensure uniformity of contamination of the clothing surfaces in this study, the surfaces were contaminated in the controlled environment of a deposition chamber. If the experimental facilities allowed it, it would be interesting to have a person walk through a plume of airborne particles and hence become contaminated in a manner more applicable to a real life scenario. One could first examine the locations on the body which picked up the highest levels of contamination. The then contaminated person could move into a resuspension chamber and engage in activities typical of indoor movement, to determine the levels of contamination which resuspend from the person. Variables which could be investigated include: (i) determining the body locations which provide the highest levels resuspension (ii) investigating how far the resuspended particles would travel into the test room and (iii) determining which indoor activities result in the highest levels of particle resuspension. A further addition to the study could be the introduction of a second 'clean' volunteer, who could then enter the test room and also perform typical indoor activities, with and without the contaminated person still present in the room. It would be particularly interesting to alter the second person's activity type to establish which is the more prominent exposure route: deposition of airborne particles (resuspended to the air by the first, contaminated volunteer) to the second volunteer as a result of walking or standing in the test room, or contact transfer of surface residing particles (deposited to room surfaces following resuspension from the first, contaminated person) as a result of sitting at a desk etc.

In terms of the contact transfer study, further detailed examination of the transition pressure area is warranted, to determine if indeed there is a step transition with a critical pressure or a sloped transition between the two pressure regimes, and also if there is a second (or more) transition pressure at higher pressure values. Additionally, an investigation using monodisperse particles of sub- and super-micron sizes would allow an assessment of the impact of particle size on transfer efficiency.

5 References

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Appendix A: APS Description and Comparison

The primary aerosol measuring instrument used in this study was the Aerodynamic Particle Sizer (APS) [Model 3321 by Trust Science Innovation (TSI)] shown in Figure 1. The APS sizes particles in the range from 0.5 to 20 μm using a time-of-flight technique, where a particle passes two laser beams that allows for the calculation of its aerodynamic diameter, and also measures the time for the particle to pass the laser allowing for coincidence detection. However, the highest recommended particle concentration is 1,000 particles per cm^3 at 0.5 μm (for <5 % coincidence) due to the flow rate, time of flight technique and electronic processing time.



***Figure 1. Aerodynamic Particle Sizer (APS).
[Picture courtesy of Trust Science Innovation (TSI)]***

Aerodynamic diameter is defined as the diameter of a sphere with a density of 1000 kgm^{-3} , which has a settling velocity equal to that of the particle in question. It is an important aerosol size parameter because it determines the particle's behaviour while airborne, regardless of its physical size, shape, density, or composition. Aerodynamic diameter was the parameter of choice in these experiments as it allows one to determine aspects of:

- Particle transport - how long the particle will remain airborne, and
- Exposure risk - if and where the particle will be deposited in the human respiratory tract

Throughout these experiments, the size of the airborne particles in both the deposition and resuspension chambers were continuously measured in real time using the APS's.

This allowed for an assessment of any shift, relative to the deposited particles, in size distribution of the resuspended particles. Within the resuspension chamber, the ‘Bottom APS’ was located at ankle height (15 cm from the floor) and the ‘Top APS’ was located at waist height (90 cm from the floor). In order to compare the data collected between the two APS’s it is necessary to compare the distributions detected by each instrument. This was achieved by the following experiment:

Both APS’s were placed into a laboratory with an open window and occasional human use. The inlets of both APS’s were connected to a single sampling tube using a y-connector. The lengths of both tubes were identical and as short as was feasible. Both APS’s were turned on and started sampling at the same time. Sampling commenced at 14:10 on 11/04/11 and stopped at 10:24 on 14/04/11, thus they were continuously sampling every second for a total of 44 hours and 14 minutes and under varied environmental conditions. The total number concentration detected in each size range was averaged over the total sampling time, for each APS and are compared in Figure 2. The horizontal axis is represented in log scale as the greatest differences are seen at the lower values of particle size.

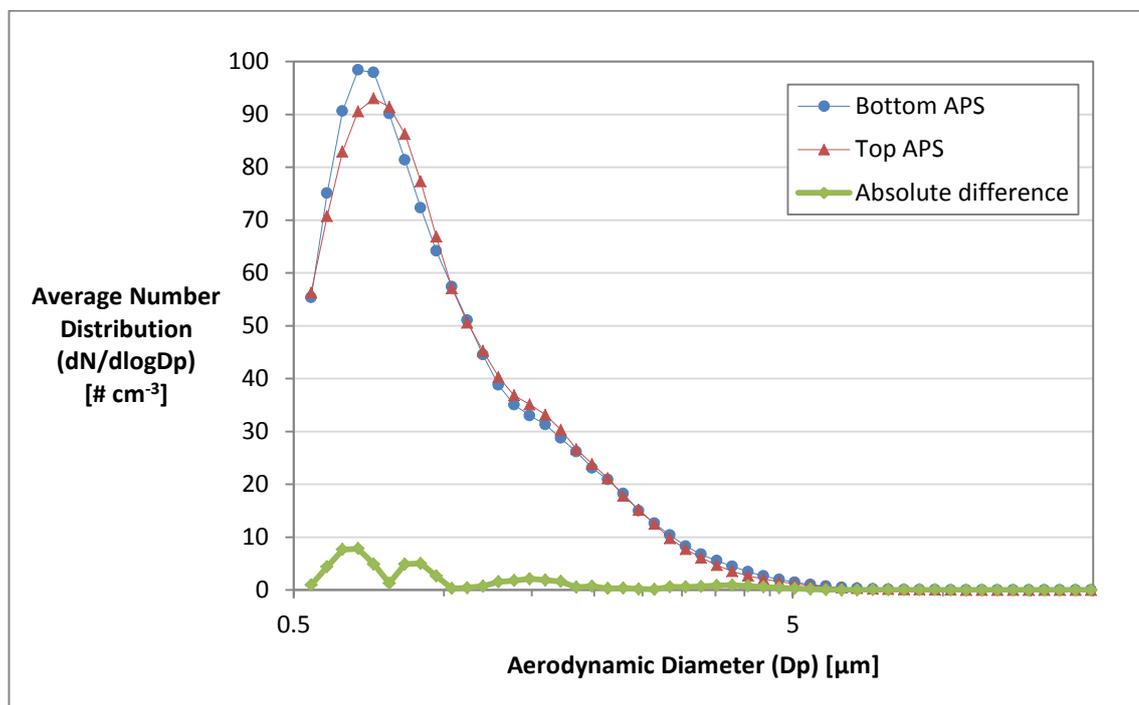


Figure 2. Comparison of the size distributions of airborne particles detected by two APS’s

As the number concentration of airborne particles increases, the difference in the number of detected particles between the APS’s also increases. This is represented in

figure 3, where the number concentration of particles detected by each APS is compared and plotted alongside a 1:1 line. It can be seen that the points deviate further from the 1:1 line as the number concentration increases. This is to be expected due to the mechanism by which the APS operates. As the APS uses laser diffraction to detect and size the particles, the higher the concentration of particles entering an APS, the greater chance of particle coincidence and hence the less reliable the APS detection will be. The manufacturers guidelines recommend concentrations of less than 1000 particles/cm³ at 0.5 µm for <5 % coincidence, and for the experiments conducted in this thesis, the highest number concentration detected was typically 30 particles/cm³, which occurred for particles of ~3 µm. Therefore it can be concluded that the data sets collected from the two APS's, are comparable.

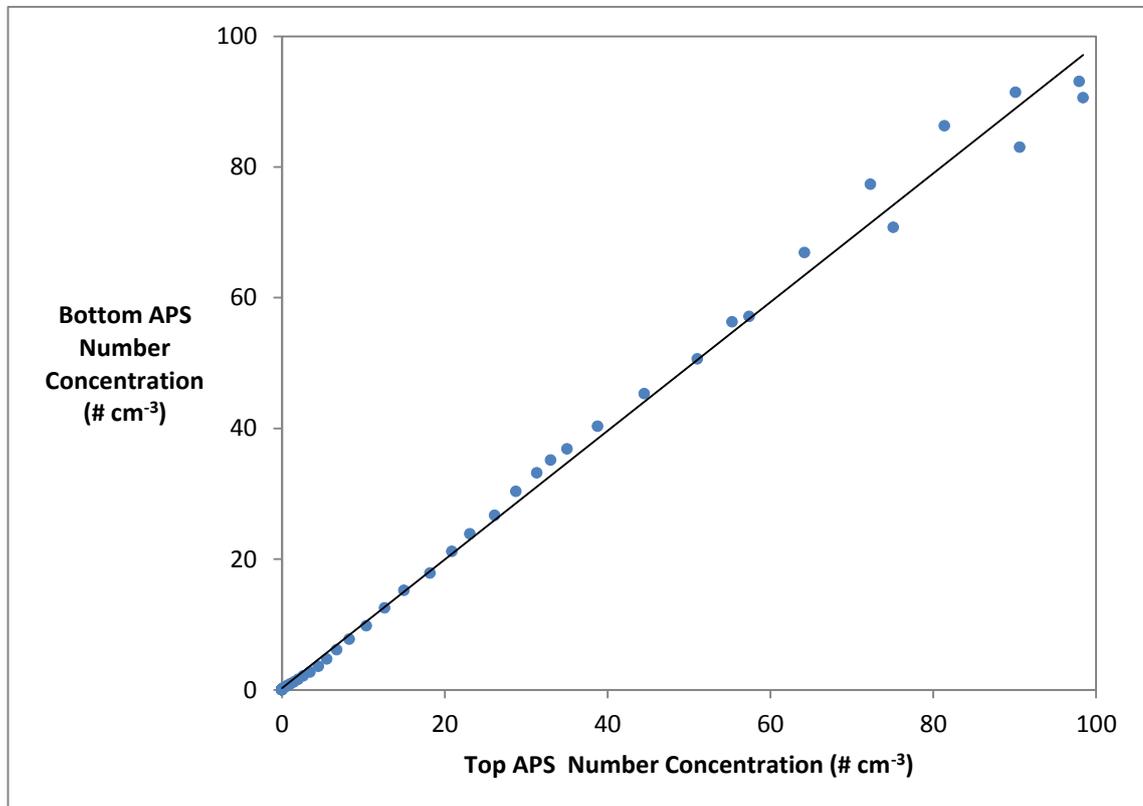


Figure 3. Comparison of the detected concentrations of airborne particles between two APS's