

*Original Article*

# Deposition and coagulation of aerosols from household spray products

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**Abstract**

This study investigated the dynamics of aerosols emitted from household spray products in terms of deposition rate, deposition velocity, and coagulation. Thirty brands of spray products were selected with respect to their uses. Time variations of particle size distribution and concentrations resulting from aerosol emission from the spray products were measured using the MetOne™ GT 321 particle counter. Deposition rates were estimated as a function of air exchange rate. Coagulation rates of singlet, doublet, and triplet particles were estimated using Smoluchowski's equation. Average deposition rates for 0.3, 0.5, 1.0, 2.0, and 5.0  $\mu\text{m}$  aerosols were 0.154, 0.157, 0.159, 0.165, and 0.170  $\text{min}^{-1}$ , respectively. The results provide useful information about the fate of aerosols released from the spray products and can be used in indoor air quality modeling and exposure assessment studies.

**Keywords:** aerosols, coagulation, deposition, indoor air quality, spray products

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**1. Introduction**

The use of household spray products leads to an increase in the levels of aerosols in the indoor atmosphere (Adeniran, Jimoda, & Sonibare, 2014; Adeniran, Sonibare, & Jimoda, 2015). The continuous presence of these airborne aerosols in indoor environments may have adverse health effects on the occupants who spend a larger percentage of their time in the indoor spaces. The dynamics of aerosols in terms of coagulation and deposition are important indicators for exposure assessment. Deposition and coagulation processes influence airborne particle concentration and size distribution (Anand, Mayya, Yu, Seipenbusch, & Kasper, 2012). These mechanisms were found to be major determinants

on the fate of indoor aerosols (Yu, Koivisto, Hämeri, & Seipenbusch, 2013). Particle deposition in the indoor environment has been recognized as a dominant mechanism responsible for a reduction in particles suspended in indoor air while coagulation results in the collision of particles of similar and different sizes leading to merging and development into larger particles (Han, Hu, & Qian, 2011; Morawska, 2005).

Estokova and Stevulova (2012) pointed out that particle deposition on indoor surfaces is governed by the processes of particle diffusion toward the surfaces and strongly depends on particle size. The major phenomena associated with aerosol deposition are Brownian and gravitational deposition (Geng, Park, & Sajo, 2013). Other factors that may influence indoor aerosol distribution include the presence of surface charge, temperature gradient, thermophoretic deposition, and room volume (Henry, Minier, Lefevre, & Hurisse, 2011).

Deposition is also an important factor in determining the degree of dermal exposure and the inhalation of the airborne aerosol particles (Shi, Li, & Zhao, 2014). Various

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respiratory diseases have been linked to the inhalation of aerosol particles (Azuma, Uchiyama, Uchiyama, & Kunugita, 2016). The pulmonary toxicity of ultrafine particles was demonstrated in a controlled laboratory experiment (Oberdörster, Oberdörster, & Oberdörster, 2005). Aerosols of diameter smaller than 2.5  $\mu\text{m}$  were linked to respiratory irritation and reduced lung function among other health outcomes (Brauer, Hirtle, Lang, & Ott, 2000). Wong *et al.* (2009) pointed out that indoor air pollution negatively influences children's health and could be responsible for sick building syndrome. Information about the fate of aerosols emitted from spray products use in the developing countries is scarce. The present study investigates the fate of aerosols released from the use of spray products in the indoor environment with respect to particle size distribution in terms of coagulation and deposition mechanisms.

## 2. Materials and Methods

This study was conducted in an occupied and unfurnished room (2.0 $\times$ 2.0 $\times$ 2.5 m) located in an uninhabited building in Ilorin, Nigeria. There were no indoor/outdoor generating sources aside from the aerosols being released from the spray products. The room was cleaned after each experimental run and the residual particulate matter measured. Spray products were applied at 1.5 m above the ground while the measuring device was placed at 1.0 m above the ground (Figure 1). The room temperature was measured when the experiments were carried out and ranged 25-30  $^{\circ}\text{C}$ , while the relative humidity ranged 50-65%. Thirty popular brands of household spray products (HSPs) were selected for the study which included 15 brands of spray insecticides and 15 brands of spray air fresheners that were available in the Nigerian market.

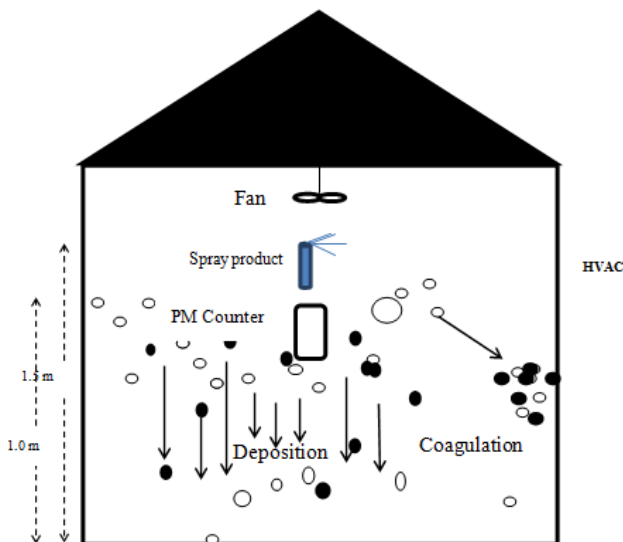


Figure 1. Description of the experimental set up.

Indoor particle deposition rate was estimated with respect to time and air exchange rate described in a previous study (Adeniran *et al.*, 2015) as:

$$\frac{dC_{ia}}{dt} = P\alpha C_{oa} + \frac{G_r}{V_{in}} - (\alpha + \kappa)C_{ia} \quad (1)$$

where  $C_{ia}$  is the indoor airborne concentration of aerosol and  $C_{oa}$  is the outdoor aerosol concentration.  $P$  is the penetration efficiency,  $\alpha$  is the air exchange rate,  $k$  is the deposition rate,  $G_r$  is the indoor particle generation rate,  $t$  is time, and  $V_{in}$  is the efficient volume of the indoor space. Since we assumed no other indoor aerosol sources except the spray from the products, Equation 1 becomes:

$$\frac{dC_{ia}}{dt} = P\alpha C_{oa} - (\alpha + \kappa)C_{ia} \quad (2)$$

To estimate the aerosol deposition rates ( $k$ ), Equation 2 was simplified by assuming that  $\alpha$  and  $k$  are constants and  $P$  equals one. Wallace (1996) gave the penetration efficiency of both fine and coarse particles to be close to unity. The time-dependent solution to Equation 2 becomes:

$$\ln\left(\frac{C_{ia}}{C_{ia_0}}\right) = -(\alpha + \kappa)t \quad (3)$$

where  $C_{ia_0}$  is the peak concentration of indoor aerosol released.  $k$  is the deposition rate which was determined by fitting a line to a plot of the log of  $C_{ia}/C_{ia_0}$  against time and subtracting  $\alpha$ , the air exchange rate, from the slope. Air exchange rate was estimated using the method proposed by the American Society of Heating, Refrigerating, and Air-Conditioning Engineers, Inc. (ASHRAE, 2010) as:

$$Q = A [a\Delta T + b(WV)^2]^{\frac{1}{2}} \quad (4)$$

where  $Q$  = air infiltration rate ( $\text{m}^3\text{h}^{-1}$ ),  $A$  = the total effective leakage area of the building ( $\text{cm}^2$ ),  $a$  = stack coefficient ( $\text{m}^6\text{h}^{-2}\text{cm}^{-4}\text{K}^{-1}$ ),  $b$  = wind coefficient ( $\text{m}^4\text{s}^2\text{h}^{-2}\text{cm}^{-4}$ ),  $\Delta T$  = average inside-outside temperature difference (K), and  $WV$  = outdoor mean wind velocity ( $\text{m s}^{-1}$ ). The coefficients  $a$  and  $b$  are 0.00188 and 0.00413, respectively.

$$\alpha = \frac{A [a\Delta T + b(WV)^2]^{\frac{1}{2}} \times 60}{R_v} \quad (5)$$

where  $\alpha$  = air change per hour, and  $R_v$  is the room volume (area of the room  $\times$  height of the room).

Aerosol number concentrations were measured using a Met One particle counter (Model GT-321, Met One Instruments Inc., USA). The equipment is a handheld, battery operated instrument that is a completely portable unit that measures five particulate number concentration ranges:  $\text{PM}_{0.3}$ ,  $\text{PM}_{0.5}$ ,  $\text{PM}_1$ ,  $\text{PM}_2$ , and  $\text{PM}_5$ . It has a sampling period of 2 min and a flow rate of 2.83 L/min. Aerosol concentrations were measured when the sampler was 1 m above ground level.

Aerosol deposition velocities were determined using the method given as (Estokova & Stevulova, 2012):

$$V_{dep} = \frac{J}{n_o} \quad (6)$$

where  $V_{dep}$  is the deposition velocity (m/s),  $J$  is the deposition flux (number deposited/ $\text{m}^2\text{s}$ ), and  $n_o$  is the uniform initial

concentration of particulate (number/m<sup>3</sup>). The influence of different ventilation scenarios on deposition of aerosol released was investigated. Different ventilation scenarios were investigated: (i) minimum ventilation (windows closed); (ii) all windows open; (iii) mechanical ventilation (fan on); (iv) mechanical ventilation (air conditioner [AC] on); and (v) mechanical ventilation (fan + AC on). The effects of smooth and rough surfaces on aerosol deposition were also investigated using smooth and rough surface carpets in the experimental room.

Smoluchowski's Equation (equation 7) was solved using the exact method (Holthoff, Egelhaaf, Borkovec, Schurtenberger, & Sticher, 1996; Menkiti, Anelke, Ogbuene, Onukwili, & Ekumankama, 2012) to estimate the coagulation to form singlet, doublet, and triplet particles.

$$\frac{N_m(t)}{N_0} = \frac{\left[\frac{t}{2(KN_0)}\right]^{m-1}}{\left[1 + \frac{t}{2(KN_0)}\right]^{m+1}} \tag{7}$$

Also, equation 7 can be expressed as:

$$\frac{N_m(t)}{N_0} = \frac{\left[\frac{t}{\tau}\right]^{m-1}}{\left[1 + \frac{t}{\tau}\right]^{m+1}} \tag{8}$$

Equation 8 gives a generic solution of Smoluchowski's equation for particle size m<sup>th</sup> order. Thus, for singlet particles (m=1):

$$N_1 = N_0 \frac{1}{\left[1 + \frac{t}{\tau}\right]^2} \tag{9}$$

For doublet particles (m=2)

$$N_2 = N_0 \frac{\left[\frac{t}{\tau}\right]^1}{\left[1 + \frac{t}{\tau}\right]^3} \tag{10}$$

For triplet particles (m=3)

$$N_3 = N_0 \frac{\left[\frac{t}{\tau}\right]^2}{\left[1 + \frac{t}{\tau}\right]^4} \tag{11}$$

where  $N_0$  = initial concentration of particles within the elemental space in question (room),  $m$  = particle size m-th order,  $t$  = particles coagulation time,  $k$  = particle coagulation rate constant,  $\tau$  = half of initial particle number/ concentration,  $N_1$  = singlet particle number/ concentration,  $N_2$  = doublet particle number/ concentration, and  $N_3$  = triplet particle number/concentration. A MATLAB M- file code was written to generate results for singlet, doublet, and triplet particles. The summation was also calculated and graphs of the number of particles were plotted against time to show the coagulation of the different particle diameters with time.

### 3. Results and Discussion

#### 3.1 Deposition of particles

The deposition trend of the HSP samples was observed from the graph. Various degrees of deposition were observed from different HSP samples. Examples of graphs are shown in Figure 2. On average, the deposition rates for insecticides were 0.155, 0.158, 0.160, 0.164, and 0.166 min<sup>-1</sup> for 0.3, 0.5, 1.0, 2.0, and 5.0 μm aerosols, respectively (Figure 4). For air fresheners, the range of deposition rate for 0.3, 0.5,

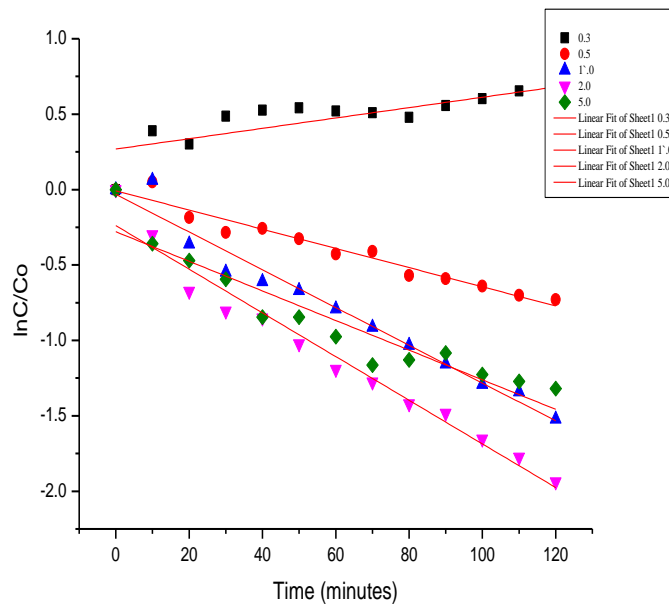


Figure 2. Deposition of aerosols from spray products with time.

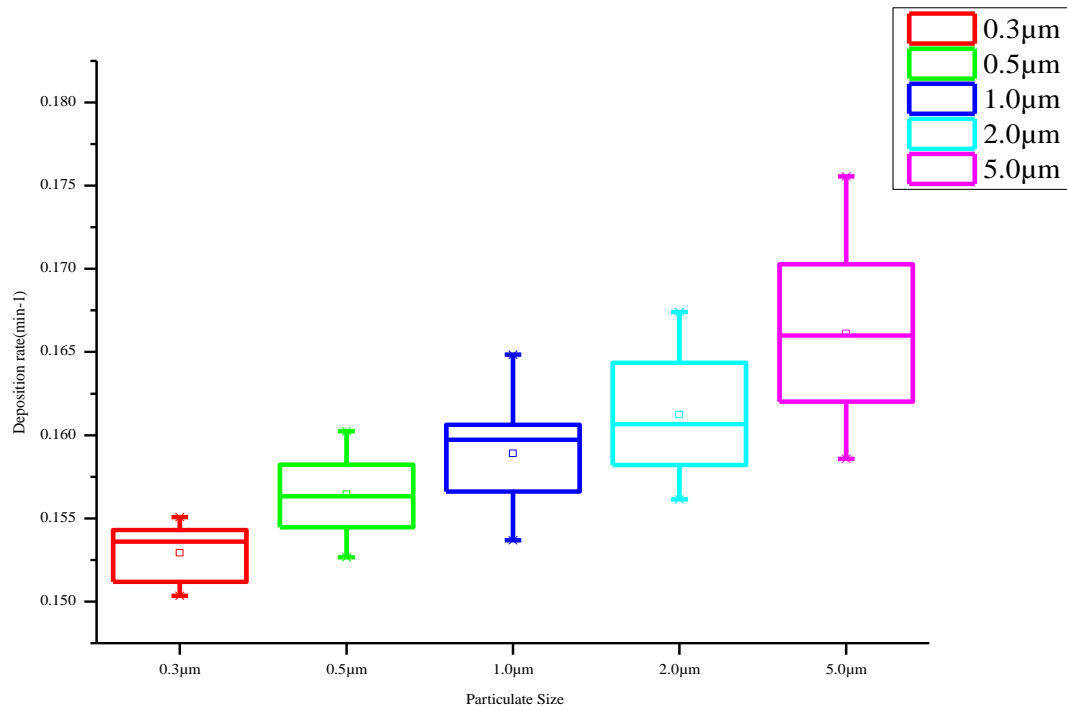


Figure 3. Deposition rate distribution of emitted aerosols from insecticides.

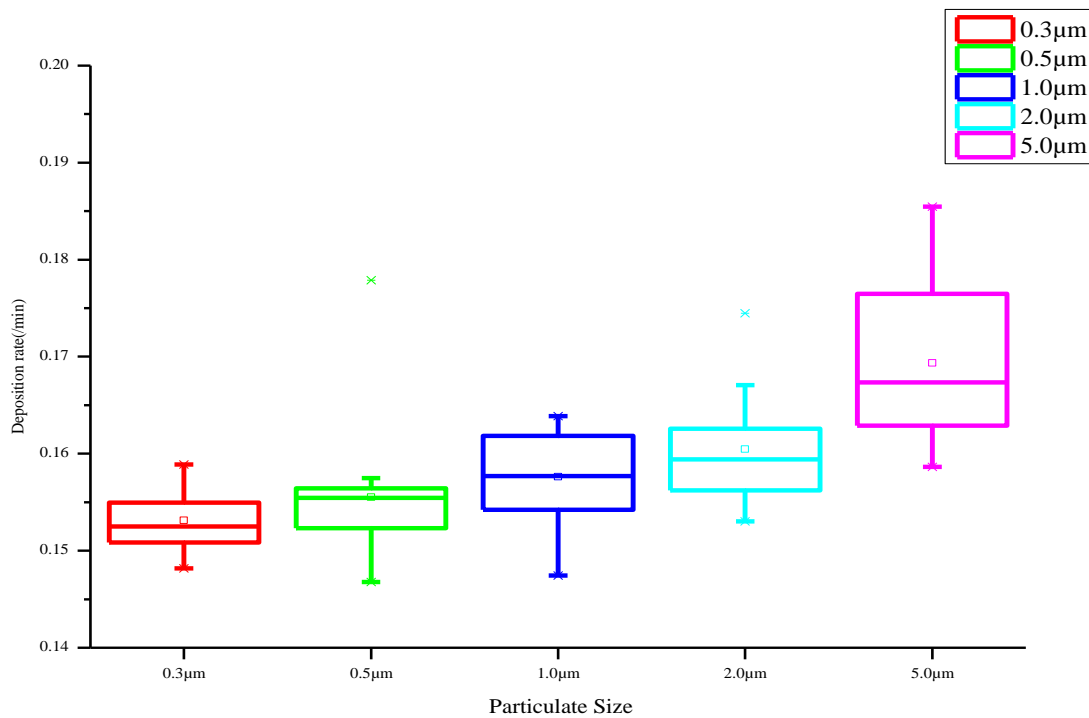


Figure 4. Deposition rate distribution of emitted aerosols from air fresheners.

1.0, 2.0, and 5.0 µm aerosols were 0.148–0.159 min<sup>-1</sup>, 0.147–0.178 min<sup>-1</sup>, 0.147–0.164 min<sup>-1</sup>, 0.153–0.174 min<sup>-1</sup>, and 0.159–0.186 min<sup>-1</sup>, respectively. Their respective average deposition rates were 0.154, 0.157, 0.159, 0.165, and 0.170

min<sup>-1</sup> for 0.3, 0.5, 1.0, 2.0, and 5.0 µm aerosols, respectively (Figure 4). The deposition rates of the particulates with higher diameters were higher than the lower diameter particles.

Previous studies on deposition rates on surfaces and coagulation of aerosol particles in a chamber concluded that particles colliding with a wall have the probability of getting stuck. Gravitational sedimentation was dominant in the deposition of particles that were 5  $\mu\text{m}$  and larger (Abadie, Limmam, & Allard, 2001; Crump & Seinfeld, 1981; Hussein *et al.*, 2009a; Hussein *et al.*, 2009b). The deposition rate of aerosol particles is determined indirectly by monitoring the concentrations of airborne aerosol particles or by directly measuring the amount of deposited particles on indoor surfaces (Hussein *et al.*, 2009). However, a direct relationship was established for aerosol size and falling time (Morawska, 2005).

Nomura, Hopke, Fitzgerald, and Mesbah (1997) reported that particle deposition in indoor air is due primarily to turbulent diffusion to the boundary layer at macroscopic surfaces in the indoor environment. Because of particle deposition, the concentrations very close to a wall surface were considered to be zero. Particles in the higher concentration areas moved to the lower concentration area by diffusion. By continuous diffusive deposition, the particle concentration of the aerosol continuously decreases. This is in tandem with the theory of particulate matter deposition by diffusion as particulates with higher diffusivity will have greater deposition rates.

### 3.2 Effects of deposition surface on aerosols

The obtained deposition rate ranges and mean values for a rough surface were 0.165–0.172  $\text{min}^{-1}$ , 0.167–0.183  $\text{min}^{-1}$ , 0.169–0.187  $\text{min}^{-1}$ , 0.177–0.191  $\text{min}^{-1}$ , and 0.176–0.194  $\text{min}^{-1}$  for 0.3, 0.5, 1.0, 2.0, and 5.0  $\mu\text{m}$  aerosols, respectively (Table 1). For a smooth surface, the obtained deposition rates ranges and mean values were 0.148–0.151  $\text{min}^{-1}$ , 0.147–0.156  $\text{min}^{-1}$ , 0.147–0.154  $\text{min}^{-1}$ , 0.153–0.157  $\text{min}^{-1}$ , and 0.161–0.167  $\text{min}^{-1}$  for 0.3, 0.5, 1.0, 2.0, and 5.0  $\mu\text{m}$  aerosols, respectively. Aerosol depositions on the rough surfaces were higher than the smooth surfaces (Table 1).

The results were in agreement with previous research studies (Abadie *et al.*, 2001; Hussein *et al.*, 2009a; Hussein *et al.*, 2009b; Zhao & Wu, 2007) which supported the assertion that there is increased deposition of particulates on rough surfaces compared to smooth surfaces. However, it can be deduced that the deposition rate of particulates increases as their diameter increases. Finer particles had a lower deposition velocity and hence a lower deposition rate (Thatcher & Layton, 1995).

Table 1. Average deposition rates of aerosols for two different types of surfaces.

| PM sizes ( $\mu\text{m}$ ) | Deposition rate for rough surface ( $\text{min}^{-1}$ ) | Deposition rate for smooth surface ( $\text{min}^{-1}$ ) |
|----------------------------|---|--|
| 0.3                        | 0.1646  | 0.1540   |
| 0.5                        | 0.1757  | 0.1531   |
| 1.0                        | 0.1773  | 0.1568   |
| 2.0                        | 0.1775  | 0.1594   |
| 5.0                        | 0.1760  | 0.1587   |

### 3.3 Effects of ventilation modes on particulate deposition

To show the importance and impact of ventilation modes on the rate of particulate deposition in real homes, the experiments were conducted in an experimental room using five different ventilation modes. The ventilation modes considered natural ventilation and mechanical ventilation. Table 2 presents the average deposition rates obtained for the different ventilation modes, i.e. minimum ventilation (windows closed), all windows open, mechanical ventilation (fan on), mechanical ventilation (AC on), and mechanical ventilation (fan + AC on).

As expected, the deposition for each ventilation mode increases with increasing size for the particulate size range of 0.3–2.0  $\mu\text{m}$  except for the minimum ventilation condition where the deposition rate of 0.5  $\mu\text{m}$  particles was slightly lower but close to what was obtained for the 0.3  $\mu\text{m}$  particles. The lower deposition rates obtained for 5  $\mu\text{m}$  particles in most of the ventilation modes were attributable to the volatile nature of the aerosols as the particulate sizes of aerosols decrease over time. Under minimum ventilation, the average deposition rate varied from 0.1531 to 0.1594  $\text{min}^{-1}$  for particulate size range 0.3–2.0  $\mu\text{m}$  and 0.1587  $\text{min}^{-1}$  for the 5.0  $\mu\text{m}$  particles.

When all windows were open, the deposition rates were slightly lower than the figures obtained than when the windows were closed. This indicated that particles were lost due to deposition and fresh air exchange. The deposition rates obtained for the different mechanical ventilation modes (fan only) and (AC only) were close. However, lower deposition rates were obtained when both fan and AC were on except for the 5.0  $\mu\text{m}$  particles. The results were in line with a previous study which affirmed that a high ventilation condition significantly reduced the indoor particle concentration due to deposition (Quang, He, Morawska, & Knibbs, 2013).

Table 2. Average deposition rates of aerosols for different ventilation modes.

| PM sizes ( $\mu\text{m}$ ) | Deposition rate MnV ( $\text{min}^{-1}$ ) | Deposition rate AWO ( $\text{min}^{-1}$ ) | Deposition rate MVFAO ( $\text{min}^{-1}$ ) | Deposition rate MVACO ( $\text{min}^{-1}$ ) | Deposition rate MVFAC ( $\text{min}^{-1}$ ) |
|----------------------------|---|---|---|---|---|
| 0.3                        | 0.1540                                    | 0.1960                                    | 0.1638                                      | 0.1579                                      | 0.1524                                      |
| 0.5                        | 0.1531                                    | 0.2435                                    | 0.1655                                      | 0.1646                                      | 0.1600                                      |
| 1.0                        | 0.1568                                    | 0.2451                                    | 0.1686                                      | 0.1678                                      | 0.1656                                      |
| 2.0                        | 0.1594                                    | 0.2475                                    | 0.1765                                      | 0.1700                                      | 0.1693                                      |
| 5.0                        | 0.1587                                    | 0.2162                                    | 0.1574                                      | 0.1693                                      | 0.1742                                      |

MnV, minimum ventilation; MV, mechanical ventilation; AWO, all windows open; FAO, fan on ONLY; ACO, AC on ONLY; FAC, fan +AC on

**3.4 Estimation of coagulation rate**

The variations in the formation of singlet, doublet, and triplet particles are shown in Figure 5. A mathematical model was developed. For the spray products considered, it was observed that the coagulation rate of aerosols increased with size. The rate of coagulation for 0.3  $\mu\text{m}$  aerosol particles was very high as the number of singlet particles reduced while those of doublet and triplet increased. Gravitational settling

occurred for 1.0  $\mu\text{m}$  particles which caused the singlet particle to be completely removed from the system. It was observed from the graph that for particles with diameter 5.0  $\mu\text{m}$ , the coagulation rate was very low with very minimal reduction in the number of singlet particles.

The graphs obtained for the various HSP samples showed a high rate of coagulation in smaller particles and low rate in larger particles. Gravitational settling was observed and some particles were totally removed from the system.

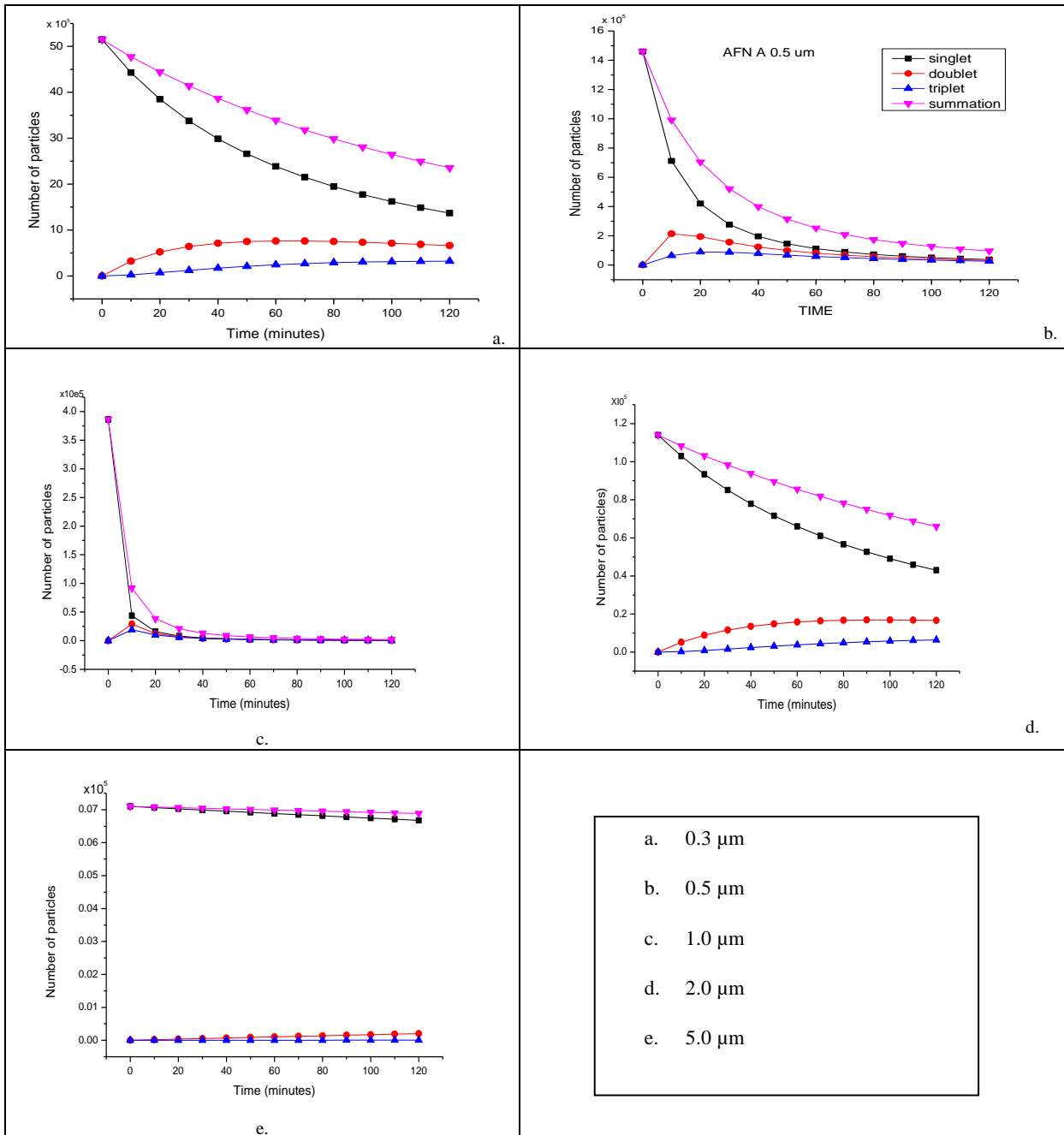


Figure 5. Coagulation of aerosols from spray products with time.

### 3.5 Deposition velocities of aerosols from spraying products

The average deposition velocities of 0.3–5 μm aerosols determined from the use of HSPs are summarized in Figure 6. The average deposition velocities of 0.3 and 0.5 μm aerosols emitted from the insecticides were 5.64E-05 and 2.04E-05 m s<sup>-1</sup>. The 1.0 μm, 2.0 μm, and 5.0 μm aerosols had deposition velocities in the ranges of 1.21E-05–9.87E-05 m s<sup>-1</sup>; 2.05E-07–9.96E-05 m s<sup>-1</sup>, and 6.70E-06–6.02E-05 m s<sup>-1</sup>, respectively.

For all the air freshener samples considered, the average deposition velocities were 9.02E-06, 1.02E-05, 1.00E-05, 4.67E-03, and 7.55E-05 m/s for 0.3, 0.5, 1.0, 2.0, and 5.0 μm aerosols, respectively.

For all 30 samples considered, the deposition velocities of the emitted aerosols were in the ranges of 1.42E-07–6.26E-04 m s<sup>-1</sup> for 0.3 μm size particles, 3.97E-07–4.23E-05 m s<sup>-1</sup> for 0.5 μm size particles, 3.78E-07–9.87E-05 m s<sup>-1</sup> for 1.0 μm size particles, 2.05E-07–6.98E-02 m/s for 2.0 μm size particles, and 2.29E-06–8.64E-04 m s<sup>-1</sup> for 5.0 μm size particles.

The average deposition velocities obtained in this study for the different aerosol sizes do not follow a particular pattern. The non-uniformity of the deposition velocities obtained from the different spray aerosols may be due to the differences in the physical and chemical characteristics attributable to their source(s). Some contain larger percentages of the total volatile organic compounds (TVOCs) while others have reduced percentages. Also, microclimatic factors and the physical conditions of the indoor environment such as temperature, air exchange rate, relative humidity, room furnishings, and airspeed could affect the deposition velocities of aerosols (Kanaani, Hargreaves, Ristovski, & Morawska, 2008; Thatcher, Lai, Moreno-Jackson, Sextro, & Nazaroff, 2002). These conditions could have various effects on the deposition velocities obtained.

However, from the obtained deposition velocities, it can be deduced that deposition velocity increases as the particulate size increases. This is in good agreement with recent studies (Lai, Wong, Mui, Chan, & Yu, 2012; You, Zhao, & Chen, 2012). The calculated deposition velocities were in the range of those obtained in previous studies. Thatcher, Lunden, Revzan, Sextro, and Brown (2003) obtained an average deposition velocity of 3.06E-04 m s<sup>-1</sup> for aerosols in the size range of 1–5 μm. Chao, Wan, and Cheng (2003) obtained deposition velocities of 1.16E-4 m s<sup>-1</sup> for 4.698–9.647 μm particles, 0.6 E-4 m s<sup>-1</sup> for 0.02–1.00 μm particles, and 0.31E-4 m s<sup>-1</sup> for 0.542–0.777 μm particles.

Dimitroulopoulou, Ashmore, Hill, Byrne, and Kinnersley (2006) obtained a deposition velocity of 3.92E-04 m s<sup>-1</sup> for PM<sub>10</sub>. Also, Tran, Alleman, and Galloo (2010) found deposition velocities of PM<sub>0.5</sub>–PM<sub>2.0</sub> and PM<sub>2.0</sub>–PM<sub>10</sub> to be 6.95E-05 and 1.94E-04 m s<sup>-1</sup>, respectively. The deposition velocities obtained can be used for modeling the deposition rates of aerosols indoors (You & Zhao, 2013; You *et al.*, 2012).

### 4. Conclusions

The present study characterizes deposition and coagulation of aerosols released from 30 household spray products. It was found that the deposition rates and velocities of the aerosols increased as the aerodynamic diameters increased. Mechanical ventilations had an influence on aerosol deposition rates as they increased the deposition rates of airborne released aerosols more than normal window ventilation. A high rate of coagulation was observed for smaller particles and low rate in larger particles. The results have provided useful information on the fate of aerosols released from spray products and can be used for indoor air quality modeling and exposure assessment studies.

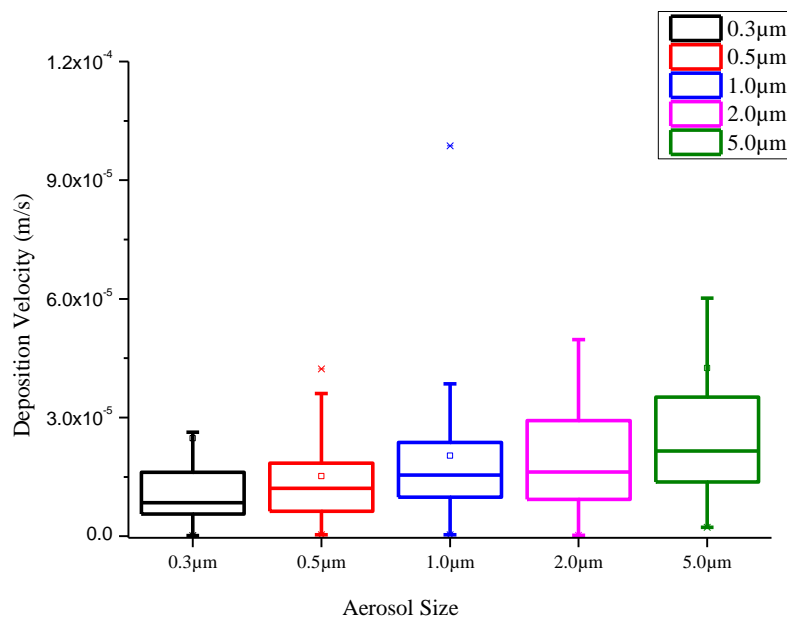


Figure 6. Average deposition velocity distribution of different aerosol sizes.

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