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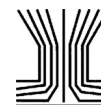
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Spray scrubber for nanoparticle removal from incineration fumes from the incineration of waste containing nanomaterials: Theoretical and experimental investigations

Emmanuel Adah^a , Aurélie Joubert^a, Rachid Boudhan^a, Marc Henry^b, Sylvain Durécu^b, and Laurence Le Coq^a

^aDepartment of Energy Systems and Environment, IMT Atlantique, GEPEA, CNRS UMR 6144, Nantes, CS, France; ^bCentre de Recherche, Séché Environnement, Saint-Vulbas, France

ABSTRACT

Nanomaterials (NMs) are currently treated via recycling, incineration and/or landfilling at their end-of-life. Little is known about the fate of NMs in incineration systems and the efficiency of the available flue-gas cleaning technologies (FGCT) in these systems on the removal of NMs before stack release. In combination with other FGCT such as cyclones, electrostatic precipitator or bag filters, scrubbers participate to limit the release of particulate matter (PM) into the atmosphere. No study has been carried out to investigate wet scrubber collection efficiency regarding nanoparticles under conditions found in a waste incineration plant. In the present study, experimental campaigns were carried out to quantify the performance of a pilot-scale spray scrubber regarding the removal of nanoparticles. The pilot was designed with respect to geometrical, hydrodynamic and residence time scale similitude and operated in gas temperature and humidity conditions representative of full-scale scrubbers in hazardous waste incineration plants. A collection efficiency of 45–62% for a particle size range of 12–90 nm was reported. To evaluate the experimental results, an existing PM collection model based on the 3 main particle collection mechanisms of diffusion, interception and impaction, was adapted for extreme humidity and gas temperature conditions typical of a waste incineration plant. A comparison of the experimental and theoretical results was made indicating that the model results were in good agreement with the experimental results. Contrary to prior studies, the impaction-dominant region occurred at smaller particle sizes (0.1–0.2 μm) corresponding to Stokes number 9×10^{-3} to 4×10^{-2} . Numerically, the contribution of the interception mechanism in the collection of nanoparticles (particle sizes 1–100 nm) was found to be negligible (i.e., Interception number 2×10^{-5} to 2×10^{-3}).

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1. Introduction

There is a growing concern about the health and environmental effects of ultrafine particles ($\text{PM}_{0.1}$) emissions (Schraufnagel 2020; Heinzerling, Hsu, and Yip 2016). Inhalable ambient particles consisting of nanoparticles (NPs) with a size range from 1 to 100 nm (Khan, Saeed, and Khan 2019) have been associated with cardiovascular and respiratory illnesses (Calderón-Garcidueñas et al. 2019a; Thomas, Al Mutairi, and De 2013; WHO 2006). The release of NPs into the atmosphere have been linked to dust cloud formation, decrease in sun light intensity and ozone depletion (Kabir et al. 2018; Smita et al. 2012). Several studies have shown that the uptake of NPs by

microorganisms and plants could lead to DNA damage, reactive oxygen species production and accumulation in the edible part of plants (Mazari et al. 2021; Sidiropoulou et al. 2018; Vittori Antisari et al. 2018). The unique properties of NPs such as size, shape and high surface area that attract their interest in industrial applications could also affect their toxicity (Kang et al. 2011). Indeed, nano-sized or nanostructured particles are more toxic than their corresponding micronic-size particles of the same chemical surface properties due to the former's increased surface area, substantial adsorption efficiency, better optical properties and increased chemical reactivity (Calderón-Garcidueñas, Reynoso-Robles, and González-Maciél

CONTACT Emmanuel Adah emmanuel-agbo.adah@imt-atlantique.fr Department of Energy Systems and Environment, IMT Atlantique, GEPEA, CNRS UMR 6144, CS 20722, 44307, Nantes, France.

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2019b; Teleanu et al. 2018; Faisal and Kumar 2017; Nurkiewicz et al. 2008; Oberdörster, Oberdörster, and Oberdörster 2005). This becomes an area of great concern when one considers the recent surge in the manufacturing and use of engineered nanomaterials (NMs) (Part et al. 2018). According to the Commission Recommendation of the EU, a nanomaterial is “a natural, incidental or manufactured material containing particles, in an unbound state or as an aggregate or as an agglomerate and where, for 50% or more of the particles in the number size distribution, one or more external dimensions is in the size range 1 nm–100 nm”(EC, 2010).

The global NMs market is projected to grow at a compound annual growth rate (CAGR) of 13.1% from 2020 to 2027 from its 2019 value of USD 8.5 billion (Grand View Research 2020). For the EU, the NMs Market is projected to reach more than \$9 billion by 2022, growing with a CAGR of 20.0% during 2016–2022 (Allied Market Research 2016). This rapid growth in the NM market has resultantly attracted increasing concerns from all stakeholders with regards to the management of NM at their end-of-life (Mishra, Arya, and Panchal 2020; Campos and López 2019; Part et al. 2018; Faunce 2017).

To date, there are no global/EU standards on the management of nanowaste. Thus, waste containing NMs are treated like any other waste, i.e., via recycling, incineration and/or landfilling without specific requirements. Research by Part et al. (2018) on the fate of NMs in commonly used waste management processes such as composting, recycling, incineration and landfilling revealed that a substantial knowledge gap exists. The authors concluded that incineration appeared to be the least investigated management process for NMs waste. Most of these studies occurred at laboratory-scale (Cernuschi et al. 2019; Ounoughene et al. 2015, 2019; Pourchez et al. 2018; Baumann et al. 2017; Buonanno and Morawska 2015; Massari et al. 2014; Buha et al. 2014; Vejerano, Holder, and Marr 2013; Vejerano et al. 2014; Derrough et al. 2013; Cernuschi et al. 2012; Mueller et al. 2012; Buonanno, Ficco, and Stabile 2009), making some of their conclusions somewhat study specific as the full complexity involved in waste incineration was not taken into account. For the large-scale studies (Oischinger et al. 2019; Baran and Quicker 2017; Börner et al. 2016; Lang et al. 2015; Walser et al. 2012), the majority of the NMs ended up in the bottom ash while some fractions of the NMs were detected in the fly ash. These conclusions were also partly supported by some of the lab-scale studies.

The resulting PM size distribution after incineration is influenced by parameters such as the nature of the specific NM, the combustion temperature (850–1100 °C), the retention time, the oxygen rate, the gas buoyancy

and the temperature gradient at the outlet of the furnace (Lang et al. 2015; Mueller et al. 2012, 2013).

To limit the release of PM into the environment from industrial processes such as waste incineration, wet scrubbers are often combined with dry scrubbing and further flue-gas cleaning technologies (FGCT) such as cyclones, fabric filters and electrostatic precipitators (Neuwahl et al. 2019).

Scrubbers are added to the FGCT primarily to treat acid gases but are also capable of handling (flammable and explosive) PM (Vallero 2019). In wet scrubbers, the absorption of acid gases is provided inside the scrubbers by pure water or a mixture of water and neutralizing additives, while PM are captured by the droplets. Spray wet scrubbers (Keshavarz et al. 2008) are amongst the most common types of scrubbers; others include: Packed bed wet scrubbers (Bhave, Vyas, and Patel 2008), Tray wet scrubbers (Schiffner 2013), Venturi wet scrubbers (Ali et al. 2013) and Gravitational wet scrubbers (Kim et al. 2001).

Particle scavenging in scrubbers is governed by several mechanisms such as Brownian diffusion, interception, impaction, thermophoresis, diffusiphoresis, centrifugal forces, condensation and electrostatic attraction. These collection mechanisms are highly dependent on the particle size distribution in the flue-gas stream. Usually, one mechanism becomes dominant for a given particle size range and acts simultaneously with other mechanisms to give a minimum collection efficiency for that particle size distribution. Contrary to wide held beliefs that conventional scrubbers are ineffective for PM < 1.0 μm, Kim et al. (2001) has shown that under favorable operating conditions PM much less than 1.0 μm can be effectively collected. Table 1 presents literature studies on the effect of operating parameters such as particle size, droplet diameters, gas temperature, gas velocity, and L/G ratio on wet scrubber performance. Only one experimental study (Vasudevan, Gokhale, and Mahalingam 1985) was identified with operating conditions close to those encountered in waste incineration regarding the droplet size and inlet gas temperature (resp. 70 μm and 200 °C). No experimental study was identified regarding nanoparticles collection.

In this study, we seek to bridge this knowledge gap by investigating the removal of nanoparticles by a pilot-scale spray scrubber designed with respect to geometrical, hydrodynamic and residence time scale similitude, and operated under realistic conditions in terms of inlet and outlet gas temperature and humidity representative of a full-scale hazardous waste incineration spray scrubber. This study is equally relevant to waste-to-energy processes such as municipal solid

Table 1. Overview of experimental and theoretical studies about particle removal by wet scrubber.

Scrubber type	Study	Numerical studies	Experimental studies (Lab- vs. Large-scale)	Particle size(μm)	Droplet size (μm)	Gas temperature (K)	Ratio liquid/gas (-)	Air/gas flow rate (m^3/h)	Liquid flow rate (m^3/h)	Fractional/Overall efficiency (-)
SWS	Mohan, Biswas, and Meikap (2008)	-	Lab	2-200	80-200	343-353	N.I.	15.79-28.58	0.03-0.12	$E_{ff} = 94.23\%$
	Tomb, Emmerling, and Kellner (1972)	-	Lab	0.68-10	225-950	N.I.	N.I.	300	0.100-0.750	$E_{ff}(\text{dp} = 0.68 \mu\text{m}) = 15\%$ $E_{ff}(\text{dp} = 2.5 \mu\text{m}) = 32\%$ $E_{ff}(\text{dp} > 3.5 \mu\text{m}) = 35\%$
	Kim et al. (2001)	Numerical approach considering impaction, diffusion and interception	-	0.1-8	100	N.I.	0.025-0.05	N.I.	N.I.	$E_{ff}(\text{dp} = 0.1 \mu\text{m}) = 9-20\%$ $E_{ff}(\text{MPPS} = 1 \mu\text{m}) = 1.8-4\%$ $E_{ff}(\text{dp} > 6 \mu\text{m}) = 100\%$
	Danzomo et al. (2012)	A new analytical approach considering Impaction	-	5 and 10 (PM10)	500-2000	N.I.	0.7 - 2.7	10.4×10^4	209.736	$E_{ff}(\text{dp} = 5 \mu\text{m}) = 43.80-99.98\%$ $E_{ff}(\text{dp} = 10 \mu\text{m}) = 58.72-100\%$ $E_{ff} = 96-99.9\%$
VWS + BCWS	Muller, Benadda, and Otterbein (2001)	-	Lab	0-400	N.I.	N.I.	N.I.	795	0.8-1.1	
	Vasudevan, Gokhale, and Mahalingam (1985)	Analytical approach considering heat transfer, mass transfer, condensation, nucleation, temperature and flow fields, and phoretic phenomena	Lab	0.1-12.3	90	653	15×10^{-4}	0.78	12×10^{-4}	$E_{ff} = 26.70\%$
	Biswas, Rajmohan, and Meikap (2008)	-	Lab	2-200	80-200	343-353	$15.9-108.1 \times 10^{-4}$	$11.1-20.1$	$30-120 \times 10^{-3}$	$E_{ff} = 50.3\%$ $E_{ff} = 60\%$ $E_{ff} = 36.7\%$ $E_{ff} = 61.5\%$ $E_{ff} = 70.5\%$ $E_{ff} = 82\%$ (hydrodynamic study)
	Centner, Büttner, and Ebert (1989)	Analytical models considering inertial impaction, turbulent diffusion and coalescence induced by turbulence	Lab	N.I.	25	293	N.I.	1000-2000	0.075-0.3	$E_{ff}(\text{dp} = 0.5 \mu\text{m}) = 20-62\%$
	Lim, Lee, and Park (2006)	Numerical approach considering impaction, interception and Brownian diffusion	-	PM10	100-2000	N.I.	N.I.	300.6	0.0468-0.2052	$E_{ff}(\text{dp} = 0.5 \mu\text{m}) = 40\%$ $E_{ff}(\text{dp} = 2 \mu\text{m}) = 90\%$ $E_{ff}(\text{dp} > 4 \mu\text{m}) = 95-99.9\%$
	Bianchini et al. (2016, 2018)	An existing mathematical model considering interception	Lab	PM2.5	100	323-328	N.I.	36	0.1-1	$E_{ff} > 99.9\%$ $E_{ff} = 28.25\%$
	Hu et al. (2021)	-	Lab	0.268-125 PM2.5	N.I.	289.2-289.6	N.I.	12024	1.35	$E_{ff} = 95.59\%$ to 96.81% $E_{ff} = 60.36\%$ (PM2.5)
	Bianchini et al. (2016)	-	Lab	PM2.5	40	323-328	N.I.	31	0.3	
	Zheng et al. (2018)	Analytical approach considering impaction, interception diffusiophoresis thermophoresis and diffusion	Lab	0.1-10	N.I.	323-363	N.I.	180	0.3	$E_{ff} = 89.75\%$ (PM2.5) $E_{ff} = 24-85\%$
	WWS	Elisa Achilles and Guerra (2020)	-	Lab	PM2.5	77.1-271.5	N.I.	0.10-0.43	N.I.	N.I.

(Continued)

Table 1. Continued.

Scrubber type	Study	Experimental studies (Lab- vs. Large-scale)	Numerical studies	Particle size(μm)	Droplet size (μm)	Gas temperature (K)	Ratio liquid/gas (-)	Air/gas flow rate (m^3/h)	Liquid flow rate (m^3/h)	Fractional/Overall efficiency (-)
	Krames and Blüthner (1994)	Lab	-	PM10	5-100	293	0.05-0.25	500-2500	1200	$E_{\text{ff}} = 99.2\%$
CPWS	Zhang et al. (2020)	Lab	-	PM100	N.I.	N.I.	N.I.	216-837	0.54	$E_{\text{ff}} = 99.85\%$
SPWS	S. H. Huang, Kang, et al. (2020)	-	A dimension-reduction method	1.5, 2.28, and 2.9	N.I.	N.I.	N.I.	N.I.	N.I.	$25\% > E_{\text{ff}} < 80\%$
OWS	Cho et al. (2020)	Lab	-	PM5	N.I.	N.I.	N.I.	192-270	N.I.	99.7% for $\text{PM} \geq 2.5 \mu\text{m}$, 89.4% for $1.0 < \text{PM} \leq 2.5 \mu\text{m}$, 62.1% for $0.5 < \text{PM} < 1.0$, and 36.5% for $\text{PM} \leq 0.5 \mu\text{m}$ Exp.: Uncharged conditions: 68.8% Charged conditions: 93.6% Theo.: Uncharged conditions: 17.4% Charged conditions: near 100%
EWS	Pilat, Jaasund, and Sparks (1974)	Lab	Numerical approach considering diffusion, impaction and electrostatic forces	0.05-5	50	293	590	N.I.	N.I.	Uncharged particle and droplet: 5% Charged particles: 25% Charged droplets: 70% Particles and droplets were oppositely charged: $> 99\%$ $E_{\text{ff}} > 99.5\%$
	Zhao and Zheng (2008)	-	Monte Carlo method for population balance considering impaction, interception, diffusion and electrostatic attraction	0.08-20	1000	433	200×10^{-4}	61072.56	N.I.	Maximum E_{ff} increase of 40% and 60%
	Carotenuto, Di Natale, and Lancia (2010)	-	Innovative mathematical model considering impaction, diffusiohoresis thermophoresis interception, diffusion and electrostatic attraction	0.1-5.0	100-500	298	N.I.	1700	0.5	Uncharged particles and droplets: very small removal efficiency Charged droplets: 35% Particles and droplets were oppositely charged: 91% $E_{\text{ff}} = \text{almost } 100\%$
	Ha et al. (2010)	Lab	-	0.3-0.9	192	N.I.	N.I.	1.2	0.048	$E_{\text{ff}} = 97\%$
	Su et al. (2020)	Lab	-	PM10	249.5	N.I.	0.017-0.125	180	N.I.	Maximum E_{ff} increase of 40% and 60%
	Di Natale et al. (2015)	Lab	-	0.01-0.45	290	Atmospheric temperature	0.88 and 1.15	0.170-0.222	0.195	Uncharged particles and droplets: very small removal efficiency Charged droplets: 35% Particles and droplets were oppositely charged: 91% $E_{\text{ff}} = \text{almost } 100\%$
	Bandyopadhyay and Biswas (2007)	Lab	-	9.82	102.3	305 ± 1	30×10^{-4}	22.3	0.066	Exp.: 88% (PM2.5) and 94% (PM10) Theo.: good agreement with the experimental results Exp. $E_{\text{ff}} = 29.8\%$, Theo. $E_{\text{ff}} = 28.6\%$
	Ru et al. (2017)	Lab	Empirical models considering impaction, interception, diffusion and electrostatic attraction	PM10 and PM 2.5	Mean diameter 150	295.8 ± 1.5	N.I.	N.I.	0.18	Exp. $E_{\text{ff}} = 88.5\%$, Theo. $E_{\text{ff}} = 87.2\%$ Exp. $E_{\text{ff}} = 97.3\%$, Theo. $E_{\text{ff}} = 97.3\%$
U-TS	Zhou, Zhou, and Zhu (2017)	Lab	Empirical models considering impaction, interception and diffusion	2	2000	323-333	1.5-2.5	103.2-348.3	N.I.	$0.5 < \text{dp} \leq 1 \mu\text{m}$: Eff increases from 52 to 97.5% $\text{dp} \geq 2$: $E_{\text{ff}} = 100\%$
TWS	Lee et al. (2013)	Lab	-	0.65-20	N.I.	N.I.	N.I.	307.8-457.2	N.I.	

WWS	Ali et al. (2013)	Mathematical model considering impaction	Lab	1	N.I.	N.I.	N.I.	457	N.I.	0.5 < dp ≤ 1 μm: E _{ff} increases from 67 to 98.5% dp ≥ 2: E _{ff} = 100% E _{ff} = 99.5%
	Mi and Yu (2012)	–	Lab	40 10	N.I.	N.I.	5 × 10 ⁻⁴	4000	2	E _{ff} = 95.1% E _{ff} = 92.7% E _{ff} = 80–96%
	Tsai et al. (2005)	–	Lab	0.05–0.478	N.I.	523 ± 293	1.17 1.50 1.80	10.8	N.I.	
HAS	Bal et al. (2019)	–	Lab	2.8–50 1.0	N.I.	303–305 N.I.	N.I.	2.09–4.18 N.I.	N.I.	E _{ff} = 99.91% E _{ff} = 97–98%
	Ahmed et al. (2018)	Eulerian–Lagrangian approach considering inertial impaction	–	–	N.I.	atmospheric temperature	0.00144 and 0.00173	N.I.	N.I.	E _{ff} = 79–99%
	Kim and Park (2020)	New approach based on one-dimensional mass, momentum and energy equations	–	0.47–1.5	N.I.	–	–	–	N.I.	–
WFGDS	Attaullah et al. (2020)	considering impaction Calvert and Mohebbi approach considering inertial impaction	–	PM1	20–80	N.I.	0.5 to 2	N.I.	N.I.	>65% E _{ff} < 100%
	Huang, Tsai, and Wang (2007)	Empirical approach based on Calvert's theory (Impaction)	Lab	0.05–0.478	N.I.	298	1.5	12	N.I.	Theo. E _{ff} > 75%
	Cui et al. (2017)	Fourth-order Runge–Kutta approach considering impaction, diffusio-phoresis thermophoresis and diffusion	Lab	0.01–10	600	288	N.I.	15 18 2.59	2.16	Exp. E _{ff} = 21–90% E _{ff} , PM ₁₀ = 100%
WFGDS	Wu et al. (2019)	A novel droplets swarm model + adaptation of impaction model	Lab	1–50	N.I.	N.I.	N.I.	79–129	1.5	E _{ff} , PM ₅ = 100% E _{ff} , PM _{2.5} = 83% Theo: E _{ff} = 88.37–88.9%
	Huang, Kang, et al. (2020)	A dimension-reduction method/Numerical approach	–	PM10	2000	343.15	N.I.	10000	132	Exp: E _{ff} = 88.9–91.7% E _{ff} = 10 – 100%
	Rafidi et al. (2018)	Euler–Lagrange CFD approach considering inertial impaction and Brownian diffusion	Lab	2–3.5	1100–2100	300–360	3.1–9.1	N.I.	N.I.	Theo.: E _{ff} = 47–76% Exp.: E _{ff} = 45–86%
WFGDS	Zhang et al. (2021)	–	Large	PM1, PM1–2.5, and PM2.5–10	N.I.	322–332	N.I.	1500–3000	4.5–10	E _{ff} = 45.2–28.3%
	Yang et al. (2010)	–	Lab	0.023–9.314	N.I.	313–393	15	99.5	1.05	E _{ff} = 40–85% (OST), E _{ff} = 77.2%
	Chen et al. (2018)	–	Large	0.1–100	N.I.	298	10	1.5293 × 10 ⁷ –2.2939 × 10 ⁷	N.I.	(TST), E _{ff} = 84.3% (FST), E _{ff} = 87.2%

Where N.I.: No information, E_g: Efficiency, SWS: spray wet scrubbers, VWS: venturi wet scrubbers, BCWS: bubble column wet scrubber, EWS: Electrostatic wet scrubber, TWS: turbulent wet scrubber, WFGDS: wet flue gas desulfurization scrubber, OST: Open scrubbing tower, U-TS: U-type scrubber, TST: tray scrubbing tower, FST: Flow pattern control device scrubbing tower, HAS: heat absorbing scrubber, IWS: Innovative wet scrubber, CWS: Cyclone wet scrubber, CPWS: composite wet scrubber, SPWS: sieve plate wet scrubber, OWS: orifice wet scrubber.

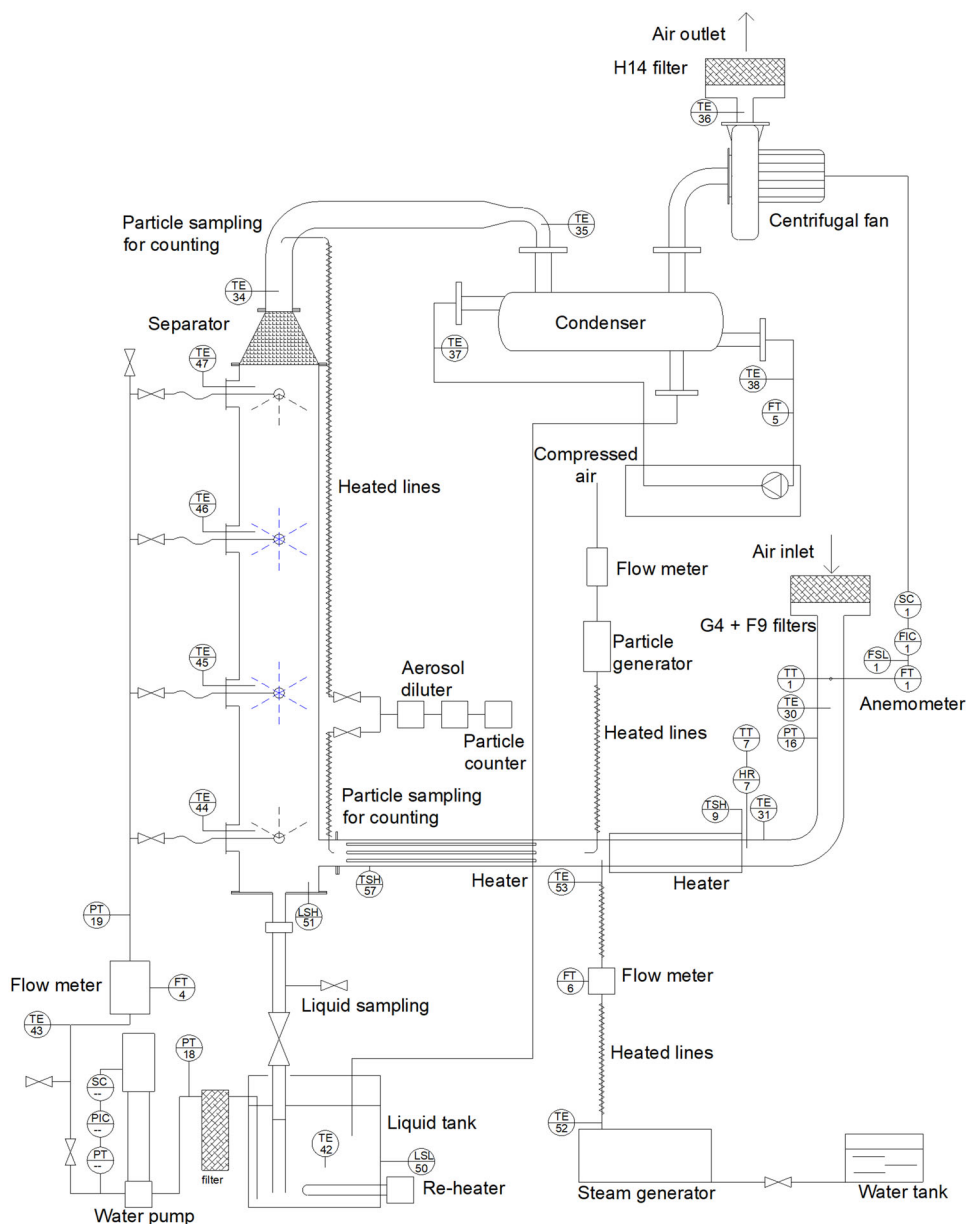


Figure 1. Schematic diagram of the set-up.

waste incineration where NMs form part of the waste mix and a wet scrubbing system is present in the flue gas cleaning line. Lastly, the experimental results are then compared to a theoretical model involving particle collection mechanisms. An investigation of the contribution of the individual particle collection mechanisms is also made.

2. Materials and methods

2.1. Experimental set-up

The principal component of the experimental set-up (Figure 1) is the spray scrubbing tower: the height is 1.9 m (excluding the mist collector), the diameter is

0.3 m. Within the tower, spray headers are located at four different stages with an adjustable number of nozzles; the configuration studied was 3-4-7-7 nozzles for headers 1 (top) to 4 (bottom) corresponding to a total liquid flow of $3.2 \text{ L} \cdot \text{min}^{-1}$. The nozzles are single-orifice with a diameter of 0.45 mm and average droplet diameter close to $75 \mu\text{m}$ (the Sauter mean diameter and the median diameters are $63 \mu\text{m}$ and $75 \mu\text{m}$ respectively) according to the manufacturer. The scrubbing tower was designed to be in scale similitude with full-scale towers encountered in the flue-gas treatment line of waste incineration plant (after ESP), in terms of inlet and outlet gas temperatures and humidity, liquid to gas flow rate ratio, ratio between height and diameter of the column, residence

time of the gas, turbulent flow regime and droplet diameter.

To operate the set-up, air from the laboratory is supplied to the set-up by a centrifugal fan (located downstream of the scrubber) after being previously filtered by a G4 and F9 filters (EN779:2012). An anemometer is used to measure the velocity flow. The airflow is conditioned in a straight length in two steps. Initially, the airflow is heated to 70 °C and then moistened by steam injection of about 9 kg.h⁻¹. Next, the airflow is further heated to 200 °C and followed by particles injection. The tests were performed with carbon NPs generated by a DNP 2000 (Palas) spark generator. The DNP 2000 generates carbon particles in the size range of 10–100 nm by spark discharge between two graphite electrodes. To evade the oxidation of the carbon, a nitrogen stream at 6 L.min⁻¹ was supplied. The carbon, evaporated in the spark, was transported by the nitrogen flow through the space between the electrodes and condenses to very fine primary particles. Depending on their concentrations, these particles coagulate to big agglomerates. A particle mass flow of 6.5 mg.h⁻¹ was generated by setting the spark frequency to 200 s⁻¹. Agglomerates were reduced by means of an exact dilution of the aerosol with clean pressured air with a volume flow of 33 L.min⁻¹.

Particle counting was performed a meter away from the generation, upstream of the spray scrubber, using a scanning mobility particle sizer (SMPS, Grimm). The scanning mobility particle sizer (SMPS) consisting of a long differential mobility analyzer (DMA) and a condenser particle counter (CPC) was employed to measure particle size distribution base on real-time selective (mobility-equivalent diameter) number concentration. The 45 particle classifications channels were used. This allowed for the possible detection of particle size range from 10–1000 nm at a sampling flow of 0.3 L.min⁻¹ requiring about four minutes for each measurement. The estimated particle concentration prior to scrubbing was 1.2×10^6 #.cm⁻³. To simplify the complex matrix of pollutants encountered in waste incineration, only NPs were generated and injected in this study.

The gas flow then made its way into the scrubber via the bottom. Water at 60 °C was used as spraying liquid. The water temperature was set to achieve the target gas outlet temperature of 70 °C. The resulting purge water was collected at the bottom in a tank, filtered and recycled back into the scrubber. At the top of the scrubber, particle counting was ensured after a mist collector. The gas flow was condensed; the

recovered water is recirculated into the water tank while the dry airflow is filtered by an H14 filter before final release to the atmosphere. To avoid the adverse effect of droplets on the particle counting by the SMPS, both upstream and downstream sampling lines were heated to 150 °C. Thus, the high relative humidity (~100%) of the gas downstream of the scrubber was reduced to 6% while the relative humidity of the gas upstream of the scrubber increased from 1% to 5%. A two-stage dilution (100x dilution factor) was then performed to lower the particle concentration, the gas temperature and humidity prior to counting with the SMPS: the first diluter was heated to 150 °C while the second diluter was operated at room temperature. In this way, stable and repeatable particle concentration and size distribution results were measured as the effects of condensation and nucleation were eliminated.

2.2. Modeling of particle collection by scrubber

The particle removal efficiency is expressed in several ways including the efficiency of a single water droplet, the efficiency of the scrubber on a mass basis, or the efficiency of the scrubber on a particle size basis. Usually, an overall efficiency of particle collection is considered. This overall efficiency considers the contributions due to the different particle capture mechanisms. In this study, we assumed that the overall collection efficiency consists of only the contributions of the three principal collection mechanisms of impaction, Brownian diffusion and interception

For particles having larger than 5 μm diameter and/or transported by gas stream velocity greater than 0.3 m.s⁻¹ (Kim et al. 2001; Perry, Green, and Maloney 1997), impaction is the dominant collection mechanism. The impaction mechanism occurs when the particles possess sufficient inertia to maintain their trajectory leaving their initial gas stream and crash with the droplet collector on their path.

Brownian diffusion is the primary mechanism responsible for collecting fine particles from a gas stream as a result of irregular motion along the gas streamline transporting the PM caused by the random collisions of the particles with gas molecules. Due to their negligible masses, the fine particles undergo diffusion movement and are captured by the liquid droplets. According to Yalamov, Vasiljeva, and Schukin (1977), the collection by Brownian diffusion mechanism occurs for particle sizes lower than 100 nm. Numerous authors (Zhao and Zheng 2008; Schnelle and Brown 2002; Pilat and Prem 1976; Johnstone and Roberts 1949) have observed that Brownian diffusion

is one of the main particle collection mechanisms in wet scrubbers.

Collection by interception occurs when a particle follows a gas streamline that is within one particle radius of the surface of the liquid droplet, it is intercepted by the liquid droplet. Interception is considered as one of the main mechanisms responsible for particle collection by water droplets in wet scrubbers for particles larger than 0.5 (Keshavarz et al. 2008; Schnelle and Brown 2002; Kim et al. 2001; Jung and Lee 1998; Gemci and Ebert 1992).

2.2.1. Collection efficiency of a single droplet, η_{SD}

2.2.1.1. Impaction, η_{imp} . To characterize the particles captured by the impaction mechanism, Stokes number (Stk) is the dimensionless parameter that translates the impaction effect and is defined as the ratio between the stopping distance of the particle and the characteristic length of the obstacle, in other words, it is the ratio of drag-to-viscous forces. The efficiency of particle removal from the gas streamlines increases as the value of the Stokes number increases. The Stokes number is defined by the following equation:

$$Stk = \frac{\rho_p d_p^2 U}{18\mu D} \quad (1)$$

Where ρ_p is the particle density kg.m^{-3} , d_p the particle diameter (m), D the droplet diameter (m), μ the viscosity of gas (Pa.s) and U the relative velocity between particles and liquid droplets (m.s^{-1}).

A mathematical correlation for the single droplet removal efficiency due to impaction was developed by Licht (1988) as:

$$\eta_{imp} = \left(\frac{Stk}{Stk + 0.35} \right)^2 \quad (2)$$

Both Seinfeld and Pandis (2006) and Kim et al. (2001) argued that when the size distribution of flue gas particles is represented by a log-normal distribution function, Equation (2) would not be suitable. The following Equations (3a) and (3b) were proposed by Kim et al. (2001) as an alternative to Equation (2).

$$\eta_{imp} = 3.4 (Stk)^{\frac{9}{5}} \text{ for } Stk \leq 0.5 \quad (3a)$$

$$\eta_{imp} = 1 \text{ for } Stk > 0.5 \quad (3b)$$

The Equations (4a), (4b), and (4c) below were developed by Lim, Lee, and Park (2006):

$$\eta_{imp} = 0.6 \cdot Stk \text{ for } Stk \leq 1.0 \quad (4a)$$

$$\eta_{imp} = 0.11 \cdot Stk + 0.49 \text{ for } 1.0 < Stk \leq 3.0 \quad (4b)$$

$$\eta_{imp} = 0.02 \cdot Stk + 0.79 \text{ for } Stk \geq 10.0 \quad (4c)$$

2.2.1.2. Brownian diffusion, η_{diff} . Pe , Peclet number is the dimensionless parameter used to describe the diffusion mechanism and is defined as:

$$Pe = \frac{DU}{D_{diff}} \quad (5)$$

D_{diff} is the diffusion coefficient of particle and is defined as:

$$D_{diff} = \frac{k_B T C_c}{3\pi\mu d_p} \quad (6)$$

Where T is the gas absolute temperature, k_B is the Boltzmann constant and C_c the Cunningham slip correction factor.

Jung and Lee (1998) developed the following expression for the diffusion collection efficiency of a single liquid droplet as:

$$\eta_{diff} = 0.7 \left\{ \frac{4}{\sqrt{3}} \left(\frac{1-\alpha}{J+\sigma K} \right)^{1/2} Pe^{-1/2} + 2 \left(\frac{\sqrt{3\pi}}{4Pe} \right)^{2/3} \left[\frac{(1-\alpha)(3\sigma+4)}{J+\alpha K} \right]^{1/3} \right\} \quad (7)$$

Where α is the solid volume fraction, σ the viscosity ratio of liquid to gas, J and K are hydrodynamic factors.

$$J = 1 - \frac{6}{5}\alpha^{1/2} + \frac{1}{5}\alpha^2, \quad K = 1 - \frac{9}{5}\alpha^{1/2} + \alpha + \frac{1}{5}\alpha^2$$

The Cunningham slip correction factor used is based on the Knudsen - Weber equation and was estimated from:

$$C_c = 1 + a \frac{\lambda}{d_p} + b \frac{\lambda}{d_p} \exp\left(-\frac{c d_p}{\lambda}\right) \quad (8)$$

Where λ is the gas molecules mean free path length; a , b , $c = 2.492$, 0.84 and 0.435 respectively (Fuchs 1964).

2.2.1.3. Interception, η_{int} . The interception parameter R , is defined as the ratio of particle diameter to the liquid droplet diameter:

$$R = \frac{d_p}{D} \quad (9)$$

R is much less than one when the droplet diameter is larger than the particle diameter. Jung and Lee (Jung and Lee 1998) defined the single droplet efficiency by interception:

$$\eta_{int} = \frac{(1-\alpha)}{(J+\sigma K)} \left[\left(\frac{R}{1+R} \right) + \frac{1}{2} \left(\frac{R}{1+R} \right)^2 (3\sigma+4) \right] \quad (10)$$

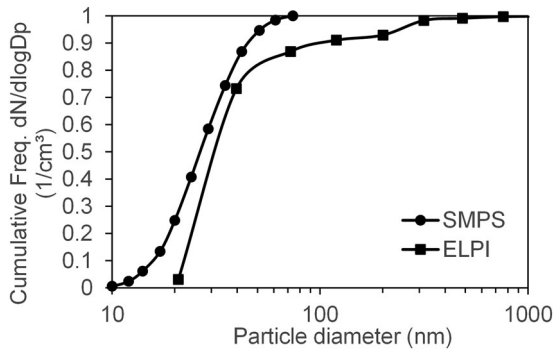


Figure 2. Cumulative particle size distribution of the carbon nanoparticles measured in the set-up upstream of the spraying scrubber with the SMPS and the ELPI particle counters.

2.2.2. Overall collection efficiency, $\eta_{overall}$

The overall collection efficiency is expressed as follows (Raj Mohan, Biswas, and Meikap 2008):

$$\eta_{Overall} = 1 - e^{\left[\frac{-3Q_L h}{2Q_G D(v_t - v_G)} \eta_{SD} \right]} \quad (11)$$

Where Q_L is the liquid flow rate ($\text{m}^3 \cdot \text{s}^{-1}$), Q_G is the gas flow rate ($\text{m}^3 \cdot \text{s}^{-1}$), v_t the terminal settling velocity of droplets ($\text{m} \cdot \text{s}^{-1}$), v_G the gas velocity in the tower ($\text{m} \cdot \text{s}^{-1}$), h the height of the tower (m).

Bearing in mind the independence of the contribution of the various particle collection mechanisms, the collection efficiency of a single droplet η_{SD} is given as (Wu et al. 2019):

$$\eta_{SD} = 1 - (1 - \eta_{imp})(1 - \eta_{diff})(1 - \eta_{int}) \quad (12)$$

3. Results and discussion

3.1. Determination of particle effective density

The elementary density of carbon is $2000 \text{ kg} \cdot \text{m}^{-3}$, however, to account for the presence of voids due to the internal structure of the particles, humidity conditions and the non-spherical nature of the carbon NPs generated by the spark generator, an estimation of the particle “effective density” was carried out. As stated by Ristimäki et al. (2002), particle effective density is not a parameter that is determined directly; it can be found if one of the following combinations is known: mobility size – aerodynamic size, mobility size – particle mass, or aerodynamic size – particle mass. Recently, a combination of particle measurements by an aerosol particle mass analyzer (APM) and a differential mobility analyzer (DMA) has been widely employed (Yin et al. 2015; Nakao et al. 2013) to determine the particle effective density.

In this study, we measured the particle size distribution (PSD) of the carbon NPs expressed with aerodynamic and electrical mobility diameters using an

electrical low-pressure impactor (ELPI, Dekati) and a Scanning Mobility Particle Spectrometers (SMPS, Grimm) respectively. The geometric mean particle diameter was 28.5 nm with a geometric standard deviation of 1.5.

DeCarlo et al. (2004) reported that, aerosol effective density (ρ_e) can be determined by simultaneously measuring the electrical mobility diameter (d_m) and the aerodynamic equivalent diameter (d_a) as shown below:

$$\rho_e = \frac{C_c(d_a)d_a^2}{C_c(d_m)d_m^2} \rho_0 \quad (13)$$

Where C_c is the slip correction factor as earlier stated and ρ_0 is the reference density ($1000 \text{ kg} \cdot \text{m}^{-3}$). We used the median diameter (D50) from the SMPS PSD with a value of 26.61 nm as d_m and the D50 from the ELPI PSD with a value of 33.38 nm as d_a (Figure 2). The effective density was found to be $1279 \text{ kg} \cdot \text{m}^{-3}$. The shape of carbon NPs are usually non-spherical chain-like aggregates (Lee 2008). However, after generation with the PALAS generator, they become loose agglomerate. Hence their effective densities decrease as the particle sizes increase (Park et al. 2003).

3.2. Particle fractional collection efficiency according to particle aerodynamic diameter

We investigated the NPs removal efficiency of the pilot-scale scrubber at typical conditions encountered in a waste incineration plant, i.e., a gas inlet temperature of 200°C , liquid flow of $3.3 \text{ L} \cdot \text{min}^{-1}$, vapor flow of $\sim 9 \text{ kg} \cdot \text{h}^{-1}$; the gas flow rate was $34 \text{ Nm}^3 \cdot \text{h}^{-1}$. Figure 3 illustrates the collection efficiency versus aerodynamic particle diameter. Due to the low resolution of ELPI in nanoparticle size classifications (Maricq, Xu, and Chase 2007), the particle size measurements were initially performed by the SMPS and the results expressed in electrical mobility diameters. Using the calculated particle effective density and Equation (13), the PSD expressed in particle mobility diameters were converted to aerodynamic diameters. The results showed a U-shaped curve for the particle size range under study (i.e., 12–90 nm) with a minimum collection efficiency of 45% at particle diameter (dp) of 35 nm. As the dp increases from 35 nm to 90 nm, a gradual increase in the collection efficiency was observed, reaching a maximum value of 62% at dp of 90 nm. Larger particles possess more inertia and hence can persist in their state of motion until they cross the fluid streamlines of the droplets. Likewise, as dp decreases from 35 nm to 12 nm, the removal of the

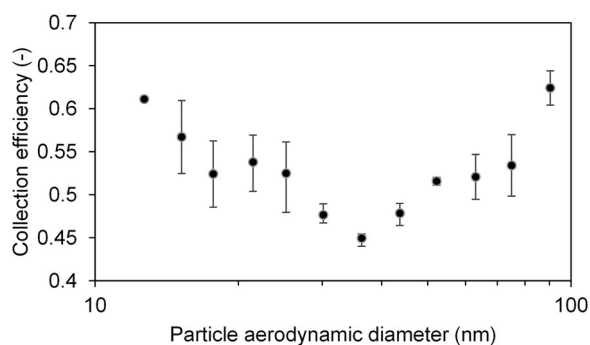


Figure 3. Experimental collection efficiency of the carbon nanoparticles by the spraying scrubber (average diameter $N=3$; range).

NPs by the scrubber increases, attaining a maximum value of 61% at dp of 12 nm. When the diameter of the particles became smaller, they experienced more random motions leading to their improved scrubbing by the droplets due to the diffusion mechanism. As stated, the minimum collection efficiency occurred at dp of 35 nm. Similar outcomes at varying operational conditions were obtained by other authors (Di Natale et al. 2015; Lee et al. 2013; Pranesha and Kamra 1996). For Lai et al. (1978) the minimum collection efficiency occurred at dp of 0.6 μm . Wang and Pruppacher (1980) noticed the same sudden decrease in collection efficiency for particles of diameter range 2 to 4 μm . Irrespective of the size range, the most penetrating particle size (MPPS) region occurs because neither of the three principal mechanisms is dominant in this region.

A point to consider is the hygroscopic characteristics of the carbon NPs and how this could affect the spray scrubber collection efficiency. Commodo et al. (2016) investigated the hygroscopic properties of organic carbon NPs and soot NPs formed in premixed flames at different carbon to oxygen (C/O) ratios and residence time. Results from static contact angle measurements by the authors revealed that the organic carbon NPs and the soot NPs from the C/O ratio of 0.67 flame were highly hydrophobic. However, the organic carbon NPs from the C/O ratio of 0.63 flame was found to be hydrophilic. The authors explained that the variation in the hygroscopic properties of the organic carbon NPs was due to the amount of surface oxygen as shown by an X-ray photoelectron spectroscopy. This surface oxygen was 6% and 3% for C/O of 0.63 and 0.67 respectively.

The carbon NPs used in the present study were generated from graphite spark discharge (i.e., graphite sublimation/aggregation at ambient temperature), as such, the C/O ratio is high. We therefore estimate

that the carbon nanoparticles have low oxygen surface function and so are rather hydrophobic.

There is currently no study on the potential effect of the hygroscopic behavior of carbon NPs on the efficiency of a spray scrubber under complex conditions such as those encountered in waste incineration plants. However, the influence of relative humidity on a venturi scrubber particle collection efficiency was investigated by Calvert, Lundgren, and Mehta (1972). Three monodisperse particles of size 0.75 μm ; pure uranine particles (PU), mixture of uranine and boric acid particles (B&PU), and Methylene blue particles (MB) were used in Calvert, Lundgren, and Mehta (1972) study. The authors observed that the PU particles grew significantly as they travel through the scrubber. Remarkably, a significant number of smaller PU particles were also observed downstream of the scrubber than those introduced upstream. Further investigations under microscope revealed that the B&PU particles also grew when exposed to varying relative humidity. However, the B&PU particles regained their initial sizes as conditions were reversed with dry air. The MB particles did not undergo size changes with changes in the relative humidity. At 0.75 μm particles, Calvert, Lundgren, and Mehta (1972) reported that the collection efficiency of MB was lowered than for both pure PU and B&PU particles. Calvert, Lundgren, and Mehta (1972) concluded that this was a result of hydrophilic particles (PU and B&PU) undergoing rapid growth as the gas contacts and atomizes the liquid in the venturi throat.

Calvert, Lundgren, and Mehta (1972) conclusions that the venturi scrubber collection efficiency of hydrophilic particles (pure uranine particles, and a mixture of uranine and boric acid particles) is higher than for hydrophobic particles (Methylene blue particles) will have to be investigated in future works for the collection of nanoparticles by spray scrubbers under waste incineration conditions.

3.3. Particle collection efficiency modeling

3.3.1. Model parameters

An existing mathematical model based on impaction, Brownian diffusion and interception phenomena was valorized for extreme humidity and gas temperature conditions typical of a hazardous waste incineration plant and the result compared with the experimental results. The single droplet contributions due to impaction, Brownian diffusion and interception mechanisms were estimated from Equations (4a)–(4c), (7), and (10), respectively. For dp of 1–100 nm and the average

Table 2. Operating parameters used for the model calculations.

Parameters	Values	Units
Scrubber height, h	1.9	m
Scrubber diameter, D_S	0.3	m
Droplet diameter, D	50–100	μm
Gas temperature, T	343	K
Droplet velocity, U_D	13.889	$\text{m}\cdot\text{s}^{-1}$
Liquid flowrate, Q_L	5.5×10^{-5}	$\text{m}^3\cdot\text{s}^{-1}$
Gas flowrate, Q_G	0.0134	$\text{m}^3\cdot\text{s}^{-1}$
Gas velocity, U_G	0.190	$\text{m}\cdot\text{s}^{-1}$
Particle effective density, ρ_e	1279	$\text{kg}\cdot\text{m}^{-3}$
Density of air, ρ_{air}	0.909	$\text{kg}\cdot\text{m}^{-3}$
Gas viscosity, μ	1.83×10^{-5}	$\text{Pa}\cdot\text{s}$
Gas molecules free mean path, λ	6.73×10^{-8}	m
Solid volume fraction, α	4×10^{-3}	Dimensionless
Terminal settling velocity of droplets, V_t	1.56	$\text{m}\cdot\text{s}^{-1}$

droplet diameter of $75\ \mu\text{m}$, the range of the dimensionless parameters of the mechanisms considered in the model are given below:

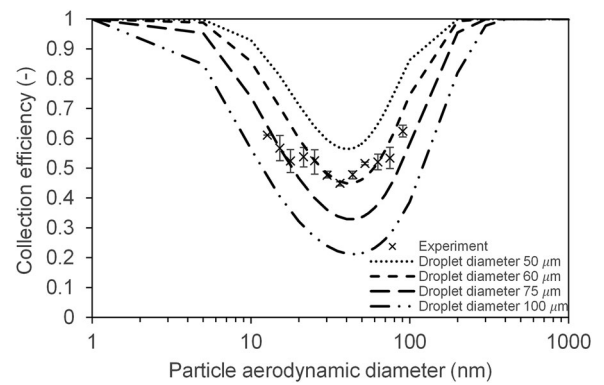
- Stokes number (Stk), 7×10^{-7} to 7×10^{-3}
- Peclet number (Pe), 2×10^2 to 1×10^6
- Interception number (R), 1×10^{-5} to 1×10^{-3}

Equation (11) was used to calculate the overall particle collection efficiency by the spray scrubber.

In Table 2 we present the parameters used for the model calculations. We calculated the droplet velocity using the continuity equation from the values of the liquid flow ($3.2\ \text{L}\cdot\text{min}^{-1}$) and internal diameter ($0.45\ \text{mm}$) of the nozzles. The packing density or solid volume fraction was determined as the ratio of the liquid volume to the tower volume. Calculating the liquid volume as the product of the liquid flowrate and the (residence) time taken for the droplet to travel from the midpoint of the tower to the base of the tower. The terminal settling velocity of droplets was calculated from the three forces acting on the droplets: drag, buoyancy, gravity forces, with the assumption that the droplets are spherical and did not undergo any form of deformation.

3.3.2. Sensitivity analysis of the droplet diameter parameter

The sensitivity of a model parameter, the droplet diameter, was studied. This parameter was studied because (i) it was not measured in the set-up (ii) a mean value was set in the model instead of size distribution. Figure 4 compares the experimental results with the results of the model at the calculated particle effective density of $1279\ \text{kg}\cdot\text{m}^{-3}$ and for different droplet diameters. Figure 4 also demonstrates the effect of the droplet diameter (D) on the particle collection efficiency. We observe a U-shaped curve with the minimum collection efficiencies occurring at particle diameter of $40\ \text{nm}$ and with

**Figure 4.** Comparison of experimental and model results.

the values of 21% for $D = 100\ \mu\text{m}$, 33% for $D = 75\ \mu\text{m}$, 45% for $D = 60\ \mu\text{m}$ and 56% at $D = 50\ \mu\text{m}$ respectively. Overall, as the droplet diameter decreased, the collection efficiency increased for the entire range of particle sizes under study. This outcome is in agreement with previous studies and can be explained by the fact that the particle-droplet contact area increases as the droplet diameter decreases.

The model results considering the average droplet diameter of $60\ \mu\text{m}$ are in good agreement with the experimental results for the particle size range studied. The better agreement at the $60\ \mu\text{m}$ droplet diameter than at the value given by the nozzle manufacturer ($75\ \mu\text{m}$) may be explained by the physical phenomenon encountered in the scrubber. As the tower gas inlet temperature is $200\ ^\circ\text{C}$ and its exit gas temperature is $70\ ^\circ\text{C}$, this will lead to a temperature gradient in the tower with certain regions with temperatures that are slightly higher than the boiling point of the droplets. This will result in the vaporization of the droplet surfaces, decreasing their sizes from $75\ \mu\text{m}$ at the exit of the nozzles to smaller diameters. This evaporation rate is moderate and its controlling parameter is the vapor diffusion rate. This is in line with studies by Aguilar et al. (2001) and Xiong and Sun (2017). We postulate that, certain regions in the tower with lower temperature (lower than $70\ ^\circ\text{C}$) and high humidity rate could lead to droplet condensation on preexisting droplets and/or particles. This evaporation/condensation phenomena could lead to a local thermal hot/cold point with reference to the gas temperature. This could also contribute to thermophoresis effects which we did not consider in the collection efficiency model.

3.3.3. Contributions of the various particle collection mechanisms

A knowledge gap observed by Kim et al. (2001), was the lack of experimental test to validate the numerous

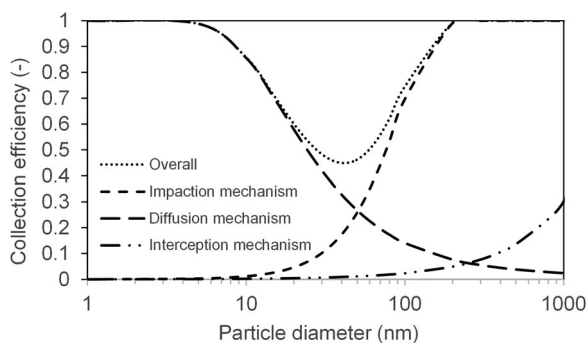


Figure 5. Contributions of the particle collection mechanisms at average droplet size of 60 μm .

theoretical studies carried out on the PM collection by wet scrubbing. As a result, prior studies have come to the conclusion that the impaction mechanism is the dominant particle removal mechanism for PM greater than 5.0 μm while Brownian diffusion is believed to be dominant for “small” PM (Lim, Lee, and Park 2006; Kim et al. 2001; Gemci and Ebert 1992; Yalamov, Vasiljeva, and Schukin 1977). As represented in our experimental results (Figure 3), the impaction-dominant region can occur at much smaller PM range. This outcome is reinforced in Figure 5, where we present the theoretical contribution of the individual particle collection mechanisms considered in this study. At dp of 0.1 μm and 0.2 μm , the collection efficiency due to impaction alone were 80.9% and 88.7% respectively. At the same dp, the contributions due to diffusion and interception were only 16.3% and 6.8%, and 2.8% and 4.4% respectively. Therefore, a better approach to determine if a mechanism is dominant or not, that accounts for the operating conditions (not only the particle sizes) of the scrubber is to use the dimensionless parameter that represent that mechanism (Equations (1), (5), and (9)). For the PM range studied (1–100 nm) and at the fitted droplet diameter 60 μm , the Stokes (Stk), Peclet (Pe) and Interception (R) numbers were 9×10^{-7} to 9×10^{-3} , 1×10^2 to 1×10^6 , and 2×10^{-5} to 2×10^{-3} respectively. Impaction was found to be dominant for Stk 3×10^{-3} to 9×10^{-3} at dp 60–100 nm, diffusion was dominant for Pe 1×10^2 to 8×10^4 at dp 1–25 nm, while the contribution due to interception mechanism was negligible. Future investigations have to evaluate the lower/upper limits of the dimensionless parameters.

4. Conclusions

In this study, an investigation of the removal of nanoparticles by a pilot-scale spray scrubber designed with respect to geometrical, hydrodynamic and residence

time scale similitude and operated in inlet and out gas temperatures and humidity conditions representative of a full-scale scrubber in hazardous waste incineration plant was carried out. The experimental results were compared to an adapted PM collection model.

Spray scrubber participate in limiting the release of nanoparticles from industrial processes such as waste incineration plants as we report a collection efficiency of 45–62% for particle size range of 12–90 nm in aerodynamic diameter, with a minimum collection efficiency of 45% at particle size of 35 nm.

The results of the model at four varying droplets sizes 50, 60, 75 and 100 μm showed that the nanoparticle collection efficiency improved as the droplet sizes decreased. The model results at average droplet sizes of 60 μm are in good agreement with the experimental results.

Contrary to prior studies that the impaction-dominant region occurs at PM $> 5.0 \mu\text{m}$, under favorable operating conditions, the impaction-dominant region can occur at much smaller PM sizes as is the case in the present study where nanoparticles collection due to impaction mechanism alone was 80.9% and 88.7% at PM sizes of 0.1 μm and 0.2 μm respectively. We therefore propose that the dimensionless parameters that represents the mechanisms, Stokes number in the case of impaction, Peclet number for Brownian diffusion and Interception number for the interception mechanism be used as the variables to consider if a mechanism is dominant or not. Future investigations have to evaluate the lower/upper limits of these dimensionless parameters.

Under the operating conditions typical of a spray scrubber in a waste incineration plant, the contribution due to the interception mechanism in the collection of nanoparticles was found to be negligible (Interception number 2×10^{-5} to 2×10^{-3} corresponding to particle size 1–100 nm). Hence, the collection of nanoparticles by the spray scrubber is dominated by impaction and Brownian diffusion mechanisms.

Declaration of conflicting interests

The authors declare that they have no conflict of interest.

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ORCID

Emmanuel Adah  <http://orcid.org/0000-0003-1902-7564>

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