Effectiveness of nanoparticle exposure mitigation measures in industrial settings

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Abstract

Inhalation of airborne nanoparticles is a well-known source of potentially health-hazardous occupational exposures. Effective mitigation measures are necessary to reduce exposure, but also challenging to implement due to the different characteristics of each individual emission source and industrial scenario. The present paper describes four different exposure case studies in the ceramic industry and quantifies the effectiveness of mitigation strategies implemented during: ceramic tile processing by thermal spraying, laser ablation, the use of diesel engines, and tile firing. The mitigation measures for exposure reduction were tailored to each industrial scenario. The NP removal efficiency of source enclosure (partial/full) combined with local exhaust ventilation (LEV) were quantified to range between 65-85% when the enclosure was partial. The efficiency reached 99% with full enclosure and vigorous ventilation (Air Change per Hour; ACH =132 h^{-1}). The elimination of the source was the optimal strategy to minimize exposure in the case of diesel forklifts use. The conventional ceramic kilns used intensively (>10 years) generated high NP exposure concentrations (>10^6 /cm^3). Appropriate maintenance and enhanced sealing enabled the reduction of exposure down to 52% of the initial value. It must be added that technologically advanced kilns, enabled even greater NP reductions (down to 84%), compared to the conventional ones. This proves technological improvements can lead to significant reduction of work exposures. This work evidences the need for tailored mitigation measures due to the broad variety of potential sources and activities in industrial scenarios. The quantitative efficiency rates reported here may be valuable for the adequate parametrization of exposure prediction and risk assessment models.

Keywords: risk assessment; worker health; hygiene and safety; non-engineered nanoparticles; ceramic industry; thermal processes

1. Introduction

The adverse effects of exposure to fine and coarse particles are well described in the literature (Pope et al., 1995; Pope and Dockery, 2006). Exposure to nanoparticles (<100 nm; NPs) in workplaces has been an issue of concern for the last decades, and the subject of numerous research studies (Brouwer, 2010; Brouwer et al., 2009, 2004; Hämeri et al., 2009; Seaton et al., 2010; Wiesner et al., 2006). The health impacts deriving from inhalation of NPs results from their capacity to penetrate into the deeper sections of the respiratory tract due to their small size has been established (Oberdorster, 2000; Oberdorster et al., 1992). NPs are also able to translocate to other body organs through the blood stream (Donaldson et al., 2005; Oberdörster et al., 2004). Other health hazardous factors are their surface area and chemical composition, which determine toxicological responses and interactions with biological molecules (Schmid and Stoeger, 2017).

Nanoparticles found in industrial workplaces and impacting exposure originate generally from two sources: (i) emission resulting from industrial activities and (ii) background aerosols. Nanoparticles in ambient background air, frequently referred to as ultrafine particles (UFPs), result from anthropogenic emissions (e.g. combustion products from vehicles) and from new particle formation (e.g., atmospheric nucleation) among other sources (Brines et al., 2015; Kulmala et al., 2014; Pey et al., 2009). The NPs emitted by industrial activities may be engineered and used as input/output in the manufacturing process, or non-engineered and formed unintentionally as a result of a given industrial activity. The latter are also referred to as process-generated (PGNPs; Van Broekhuizen et al., 2012) and incidental NPs (Viitanen et al., 2017) are the subject of the present study which is focused on ceramic industry.

A recent literature review, which assessed publications reporting industrial sources of UFPs particles and exposure concentrations in workplaces (Viitanen et al., 2017) concluded that real exposures (e.g. in welding and metal industry) were more than hundred times greater than those resulting from background aerosols. The obtained results of measurements were not conclusive enough to draw general conclusions with regard to exposure. In particular, NP release in the ceramic industry resulting in worker exposures can be found in traditional pottery (Voliotis et al., 2014), in ceramic tiles sintering (Fonseca et al., 2016) and in innovative processes (Fonseca et al., 2015; Salmatonidis et al., 2018) such as high energy ones (e.g. thermal spraying;

Salmatonidis et al., 2019; Viana et al., 2017). Hence, there is sufficient evidence to conclude that unintentional NP release generates statistically significant impacts on worker exposure in the ceramic industry.

Consequently, efficient exposure mitigation strategies must be implemented. Certain measures are based on industrial practices and on the hierarchy of control methods (Conti et al., 2008; E.U., 2014; Gerritzen et al., 2006; Schulte et al., 2008). The European Council Directive 98/24/EC (E.U., 2014) recommends the elimination or isolation of sources as methods to minimize exposures to hazardous substances in workplaces. If these measures are not applicable, engineering controls should be applied (e.g. dilution and local exhaust ventilation) and finally personal protective equipment (PPE), such as respirators or masks are recommended. A recent review that quantified the efficiency of PPE and engineering controls (Goede et al., 2018), especially for engineered NPs in controlled scenarios such as laboratories, reported that the available data are inconclusive. Previous studies discussed the efficiency of PPE such as protective gloves, clothes, filtering facepieces respirators and half mask respirators (Kim et al., 2006, 2007; Lee et al., 2007; Myojo et al., 2017; Tsai et al., 2010). A review of this literature, based on search terms “nanoparticles”, “protective equipment” “ventilation”, “extraction”,” safety”, “mitigation”, evidenced that: (i) studies for incidentally-released NPs are less frequent than for engineered ones; (ii) in spite of a great number of studies on PPE efficiency, less information can be found about the effectiveness of applied technical measures, and they refer mostly to laboratory-scale; (iii) the results obtained cannot be easily generalized beyond the specific cases; and (iv) experimental studies at industrial scale constitute a clear research gap. The diversity of industrial processes poses a major challenge when assessing the effectiveness of exposure mitigation measures. The literature shows that data regarding the efficiency of NP exposure mitigation measures in real world facilities, at an industrial scale, are scarce and not standardized. This is not the case for coarse and fine particles, for which the Exposure Control Efficacy Library (ECEL) provides information on the efficacy of control methods for inhalation exposure (Fransman et al., 2008), mainly focusing on particle mass concentration as main metric (as opposed to particle number concentration, used for NPs). It should be added that the quantitative data on exposure reduction for specific technological measures are also a key input for exposure prediction models applied to indoor settings in the framework of risk assessment (e.g., one- and two-box models; Hewett and Ganser, 2017; Hussein and Kulmala, 2008; Nazaroff, 2004; Ribalta et al., 2019).

The present work aims to quantify the efficiency of measures for NP exposure reduction implemented under real-world operating conditions in the ceramic industry. These measures include: (i) ventilation (extraction and dilution); (ii) source enclosure; (iii) source substitution; and (iv) periodical source isolation. The efficiency of the measures was assessed by a case study approach. The approach presented consists of characterization of NP exposure before and after the implementation of mitigation measures. The exposure reductions are characterized by the measurements of particle number concentrations. It should be noted that this study does not aim to discuss the measured exposure concentrations from a regulatory compliance perspective. Thus, this work aims to expand the current literature on exposure mitigation strategies by contributing with quantitative assessments of effectiveness of specific technical measures. The data obtained will make a valuable contribution for the adequate parametrization of exposure prediction and risk assessment models.

2. Materials and methods

2.1 Particle emission scenarios

Four particle emission scenarios were evaluated:

(A) *Thermal spraying deposition of coatings*

Particle monitoring was carried out during processing using atmospheric plasma spraying (APS) in a semi-industrial pilot plant. Details on this industrial technique and on the NPs generated may be found elsewhere (Salmatonidis et al., 2019; Viana et al., 2017). The APS installation was located inside the spraying room with a torch installed on a robot. The pilot plant included three compartments like the one in Figure 1. The spraying room and the worker's room were connected by an interior door (Figure 1), which may remain either closed or open (binary condition) during processing. The door remains closed in routine processing (source enclosure). Sometimes, the operator should intervene manually and the door was open (source partial enclosure). The APS area was equipped in with a local exhaust ventilation (LEV) system. The particle monitoring locations were: (i) the spraying room (emission source); (ii) the worker´s room (exposure area); and (iii) outdoor background (located in the corridor outside of the worker´s room; Figure 1). The monitoring instruments were placed on a desk, next to the operator at breathing height but not directly at the worker breathing zone. The mitigation variables modified were door configuration (closed or open) and extraction flow rate in the studied APS rooms, these two variables can be expressed in a single parameter: air changes per hour (ACH).

(B) Laser ablation of ceramic tiles

The use of this technology in ceramic tile treatment and NP release mechanisms were studied previously in laboratory (Salmatonidis et al., 2018) and in pilot-plant scales (Fonseca et al., 2015). In this case study, the NP emissions associated to laser ablation of fired ceramic tiles was studied in an industrial facility, in which the laser source was located in a partially closed chamber having volume of 5.6 m^3 equipped with a LEV system having ventilation capacity of about ca. 2000 /m^3. The laser processing was carried out discontinuously, with laser working cycle duration of ca. 2 minutes. The measurements were performed at a distance of ca. 0.5 m from the emission source what is, representative of the worker exposure area (Figure 2). The efficiency of NP reduction was measured at: (i) laser inactivity (background); (ii) laser ablation with LEV; and (iii) laser ablation without LEV.

(C) Diesel engines emissions

The machines powered by diesel engines are widely used in indoor industrial facilities (Gaines et al., 2008). The use should be reduced to comply with the upcoming indoor air exposure limit values for carcinogen contaminants such as diesel soot measured as elemental carbon set by the European Council Directive 2019/130 (EU, 2019). The directive sets the concentration limits equal to 0.05 mg/m^3 after the year 2023., The impacts of the use of two Toyota 2z forklifts having power of 42 kW (EU stage II clear) was studied inside an industrial workplace. The forklifts were continuously operating inside the plant performing loading and unloading of material pallets. In this experiment it was not possible to isolate the source from any secondary ones because of their continuous movement. However, it could be assumed that diesel forklifts were the main NPs source in the worker´s breathing zone. The particle concentration monitoring was performed in a stationary location in the loading and unloading area (worker area). Moreover, a personal monitor was worn by the forklift operator (breathing zone), working in an open cabin. The mitigation measure studied was source substitution based on the use of electrically powered forklifts instead of the diesel ones.

(D) Ceramic tile firing in a roller hearth kiln

The study was carried out in an industrial plant for production of ceramic tiles (glazed white-body earthenware wall tiles) under real operating conditions (peak temperatures around 1150ºC; Ferrer et al., 2015). The activity included the use of a roller kiln (120 m-long), which is the most frequently-used technology for firing ceramic tiles (Mezquita et al., 2014). The experimental measures were performed outside the roller kiln at 1.5m in height and 2m aside from its external walls, every 10 m along the kiln. The monitoring of NPs was performed in three areas which correspond to the firing cycle: heating, firing and cooling. Three particle monitoring campaigns were carried out in the industrial plant. The first campaign monitored a conventional kiln being in an intensive service for ca. 10 years. The second campaign at the former kiln after having done maintenance works in the refractory walls. The third campaign was carried out in a new and technologically advanced kiln with optimized refractory conditions (being less than 2 years in service).

2.2 Mitigation strategies implemented and assessed

The case studies were performed to allow the assessment of three different mitigation strategies. The strategies were classified following the hierarchy approach (E.U., 2014; Schulte et al., 2008) as:

- **source substitution/elimination,** tested in particle emission scenario (C) in which the diesel forklifts were substituted by the electric ones.
- **source isolation,** tested in particle emission scenario (D) which comprised maintenance and sealing improvement for enhancing source enclosure, during the operation of a roller kiln firing ceramic tiles.
- **engineering controls,** tested in the particle emission scenarios (A) and (B) (thermal spraying and laser ablation, respectively) in which ventilation and LEV system were combined with source enclosure.

The particle emission scenarios and the mitigation measures are shown in Table 1. All data were obtained under real industrial operating conditions. The conditions include the production scale (from kgs to tons), facility surface area (from tens to thousands m^2), the number of workers (from two to hundreds). The efficiency of mitigations measures was quantitatively determined, but some practical limitations must be mentioned. Namely, the different mitigation measures overlapped in some scenarios (e.g., LEV and partial source enclosure were operating in parallel in the APS facility) or potential influence of external sources resulting from inadequate isolation of studied areas B, C and D.

2.3 Particle monitoring instrumentation

Workplace exposure assessments were carried out by monitoring particle number concentration and their mean diameter, using online instrumentation (Table 2). The monitors measured particle diameters range from 4nm to 32µm. Particle number concentrations were monitored with fixed and portable instrumentation (TSI CPC 3775; DiSCmini, TESTO) and size distributions were measured using NanoScan-SMPS (TSI 3910) and a laser spectrometer (Mini WRAS 1371, GRIMM). All instruments were intercompared prior to the measurements for quality assurance purposes. The performance of the DiscMini and NanoScan monitors and the intercomparison methodology were recommended elsewhere (Fonseca et al., 2016; Viana et al., 2015).

Particle number concentrations were monitored at the emission source, in the worker area or in the breathing zone (depending on the scenario) in indoor and outdoor locations (OECD, 2015; Ramachandran et al., 2011). The indoor (background) location was located at a distance greater than 2 m from the emission source in each case to avoid potential interferences. The outdoor location was to evaluate the possible contribution of outdoor sources (e.g., road traffic).

The effectiveness of the exposure mitigation measures (E_{EMM}) was quantified according to Eq. 1:

$$E_{EMM} = (1 - \frac{C_{EMM}}{C_0}) \times 100 \qquad (1)$$

where C_0 is the initial particle number exposure concentration before the implementation of mitigation measure, and C_{EMM} is the concentration after its implementation.

The industrial processes do not enable to perform always the measurement without mitigation because of safety requirements. The different approach for calculating the efficiency was applied when mitigation measures were already implemented (e.g., case study A). Namely, the emissions were monitored simultaneously in the emission source and in operator area and the total reduction of particle concentration was calculated according to Eq. 2,

$$E_{EMM} = (1 - \frac{C_{WA}}{C_{ES}}) \times 100 \qquad (2)$$

where C_{WA} is the number concentration in the worker area and C_{ES} in the emissions source.

3. Results and discussion

3.1. Source substitution/elimination (C; Diesel engines emissions)

Mitigation measures: the measures implemented consisted of substitution of diesel forklifts (Toyota 2z, 42 kW, EU stage II clear) by electrical ones (STILL RX60-25, emission-free drive) to reduce indoor exposure to soot NPs.

Particle emissions: mean particle number concentrations were 1.1 $\times 10^5$/cm^3 in the worker area, with their mean diameter of 39 nm in the monitored range 10-420 nm. The peak of particle number concentrations in the breathing zone was greater than 2.5*10^3/cm^3 (1-min mean concentrations), corresponded to low mean particle diameters (30-40 nm), characteristic of diesel emissions (Kittelson et al., 2004; Morawska et al., 2008). The particle size distribution in the worker area was lower than 50 nm (83% of the particles) and 51% of them were lower than 30 nm.

Efficiency of the mitigation strategy: Figure 3 shows the comparison between particle number concentrations monitored in the breathing zone, during operation with diesel and with electrical forklift (during 1 h period). Measurements were recorded on two different days, with a time interval of one week. Only one type of forklift was evaluated on each of the days, initially the diesel and then the electrical ones. Background concentrations were monitored simultaneously in a background reference location in the plant, using a DiscMini monitor. This area was not directly affected by any process, and it was located >5m away from the forklift area. Results showed lower particle number concentrations when electric forklifts were used. The maximum exposure concentration (ca. 1*10^5/cm^3) was comparable to the lowest ones recorded when the diesel forklifts were not in operation. A reduction of 49% of particle concentration in the breathing zone, shown in Table 3, was calculated when electrical forklift was used instead of diesel one. This reduction is an average for 1 h monitoring period where forklifts were both operating and stationary. When focusing on the forklift driving intervals and by subtracting the background concentrations, the efficiency of source substitution was 92% (Table 3). It did not reach 100%, due to the fact that measurements were taken on different dates and because of the influence of secondary sources such as diesel engines working outdoors. The re-suspension of the previously deposited fine and coarse particles (with lower contributions in terms of particle number) by the electrical forklifts might have contributed as well.

3.2. Source isolation (D; Ceramic tile firing in a roller hearth kiln)

Mitigation measures: two strategies were implemented for assessing the effect of the source enclosure in scenario D: (i) kiln refurbishing by improving the sealing of 10 years old kiln; (ii) replacement of a conventional kiln by a new one of advanced technology and with optimized refractory conditions. To do so, three experimental campaigns were carried out.

Particle emissions: during the first of the three campaigns, in the conventional kiln without implementing any mitigation measure, the highest particle number concentrations were recorded in the zone of maximum temperature of the firing cycle (>8*10^5/cm^3; 46 nm; see Figure 4), which is the main emission area due to the highest temperatures recorded (Fonseca et al., 2016). Concentrations were constant over time in this region, 75% of the particles showed sizes smaller than 50 nm and 40% were smaller than 30nm, indicating nucleation as the main formation mechanism which is also consistent with the literature (Fonseca et al., 2016).

Efficiency of the mitigation strategies: as a first stage to mitigate worker exposure to high NP concentrations, the maintenance and sealing of the old kiln in the firing zone were carried out. The efficiency of these measures was evaluated during the second monitoring campaign. Additionally, in a third stage, a new high-efficiency roller kiln was installed and its emission efficiency was also assessed. The reduction of NP emissions throughout the three different campaigns was observed (Figure S1, Supplementary material). The concentration for the old kiln dropped from $1\times10^6/cm^3$ to $5\times10^5/cm^3$ for the refurbished kiln and down to $1.6\times10^5/cm^3$ measured for the new, advanced kiln (Figure S1).

The adequate maintenance of the old kiln, including enhanced sealing, resulted in 51.6% of NP concentration reduction (Table 3). The particle number concentration decreased from 1×10^6 to $5\times10^5/cm^3$ along the wall of the kiln's firing zone. The use of new kiln reduced NP concentrations by 84.4%, compared to the old and refurbished one (Table 3). The concentration decreased from $1\times10^6/\ cm^3$ to $1.6\times10^5/\ cm^3$ as shown in Figure 5. These decreases were linked to the different conditions of the refractory materials, which were used to insulate the firing compartment of the kiln. Whereas the renovation of the conventional kiln was able to reduce particle release (52%, Table 3), this reduction was lower than that obtained from the operation of the advanced kiln with superior refractory sealing and energy efficiency, which proved to be also more efficient in terms of emissions reduction (84%, Table 3). In the cooling sections of the kilns (Figure 4a), results evidenced that the exposure concentrations around both kilns (conventional and advanced) were similar. Thus, the effective as well as targeted enclosure of the firing process, the optimum refractory condition and maintenance of the insulating materials are key parameters governing workplace exposure in ceramic tile firing facilities.

3.3 Engineering controls: ventilation and LEV system combined with source enclosure (A; thermal spraying deposition of coatings, and B; laser ablation of ceramic tiles)

The efficiency of specific ventilation and LEV combined with source enclosure measures was assessed in two particle emission scenarios described above, namely during: (A) thermal spraying deposition of coatings, and (B) laser ablation of ceramic tiles.

(A) Thermal spraying deposition of ceramic coatings

Mitigation measure: a LEV system was located directly above the APS area, extracting 24000 m^3/hour from three spraying rooms. Therefore, the extraction rate of the LEV fluctuated with the number of APS installations operating simultaneously. It should be noted that the maximum extraction rates can be considered high for such a pilot plant. The LEV system included a capturing hood covering the emission source and a duct without flanges with 0.36m diameter. An open hatch on the ceiling provided air supply to the spraying room when the LEV was active. The air exchange rate (ACH in Eq. 3) varied from 132 to 33 h^{-1} depending on the extraction flow rate of the LEV system and on the binary condition of the interior door (open/closed) influencing the total volume of affected area. The ACH was calculated according to Eq. 3:

$$\frac{Extraction\ flow\ (\frac{m^3}{h})}{Volume\ of\ affected\ area\ (m^3)} = ACH\ (h^{-1}) \qquad (3)$$

Particle emissions: in total there were 30 spraying events, 11 for spraying micro-sized NiCrAlY and ZrO_2+(4mol%)Y_2O_3 powders and 19 times for spraying liquid precursors $Zn(NO_3)_2 \cdot 6H_2O$, $Zn(O_2CCH_3)_2$, $C_8H_{12}O_8Zr$). Table 4 summarises the details about representative spraying experiments, as well as the measured exposure concentrations in terms of particle number inside the spraying and the worker room. Mean particle number concentrations ranged between $3.7*10^5/cm^3$ - $1.5*10^6/cm^3$ and mean particle sizes were in the range 26-45 nm inside the spraying room, while in the operator area concentrations ranged from $3.3*10^3/cm^3$ to $5.4*10^4/cm^3$ (Table 4) and sizes from 32-59 nm. For all of the experiments, particle number concentrations were orders of magnitude higher inside the spraying room than in the operator area even when the door was open.

Efficiency of the mitigation strategies: different ventilation and door configurations were tested. The most effective mitigation configuration corresponded to the highest ACH rate (132 h^{-1}, experimental runs #2 and 3, powder feedstocks; Table 4). In these experimental conditions the door was closed and the emissions generated inside the spraying room were not transferred to the operator room (Figure 6). The particle number concentrations did not demonstrate any statistically significant increase in the worker room, for both types of powders. Such experimental conditions resulted in about 99% reduction of particle number concentrations between spraying room and the worker – exposure – area (see Table 3).

The experimental runs #1 and #2 used the same feedstock (powder) and were performed under the same enclosure conditions, while different ACH values were applied (#1: 66 h^{-1} and #2: 132

h^{-1}; Table 4). However, the efficiency of exposure reduction for the experimental runs #1 (99.3%) and #2 (98.5) were similar and approximately 99% indicating the significance of enclosure against fluctuations on the intensity (flowrate) of a continuously working LEV.

The experimental runs #4 and #5 (liquid-precursor feedstock), were carried out at ACHs of 33 and 66 h^{-1}, respectively and the interior door was open. As expected, when ACH had the lowest value (33 h^{-1}), mean exposure concentrations in the operator area had the highest value ($5.4*10^4/cm^3$; Table 4). Although, the peak concentrations were similar under both ACHs values (see Figure 7a), for ACH=33 h^{-1}, the particle number concentrations decreased at a slower rate than for ACH= 66 h^{-1} resulting in wider peaks with a higher potential for exposure impacts (Figure S2 in Supplementary material). When the air extraction rate was the highest (132 h^{-1}; Figure 7a; during spraying of powders), the peak particle number concentrations were lower than that measured during spraying of liquid precursors (lower ACH). It can be observed that the particle number concentrations decreased at a slower rate when powder was used as feedstock as opposed to liquid one, despite of ACH being almost 4 times higher (132 h^{-1} with powders vs. 33 h^{-1} with liquids; see Figure 7a). This evidenced the influence of the process parameters, as well as the technical mitigation measures implemented. Nevertheless, further research would be necessary to understand the influence of the use of powder or liquid feedstock.

In order to evaluate the influence of enclosure as mitigation measure, the experiments with the same LEV extraction rate (24000 m^3/h) and different door positions are compared in experimental runs #3 (powder feedstock) and #5 (liquid-precursor feedstock). Because of the air volumes were different when the door was open or closed, the ACH factor at the experimental run #3 was 132 h^{-1} and only half of this value, i.e. 66 h^{-1} during experimental runs #5 (liquid-precursor feedstock). During experimental run #3 (powder feedstock; ACH=132 h^{-1}) the mean efficiency of exposure reduction was 98.5%, while during experimental run #5 (liquid-precursor feedstock), with the door open and the same extraction flowrate (24000 m^3/h), the exposure reduction was 95.4% (see Table 3). It can be concluded that, for experimental runs #3 (powder feedstock) and #5 (liquid-precursor feedstock), the impact of the extraction flowrate on exposure mitigation was stronger than that of the enclosure (door open/closed). The difference in reduction efficiency becomes wider when experiments with lower extraction rate (12000 m^3/h) and different door positions are compared (#1 vs. #4; powder vs. liquid-precursor feedstock, respectively). The efficiency decreased to 85.6% during experiment run #4 (liquid-precursor feedstock), which was performed with the door open, while when the door was closed the efficiency was higher (99.3%; Table 3). According to this comparison (#1 vs. #4; powder vs. liquid-precursor feedstock, respectively) the enclosure has a higher influence in reducing exposure than the previous comparison (#3 vs. #5; powder vs. liquid-precursor feedstock, respectively), which is an indication that enclosure becomes more effective when LEV is less efficient, and vice versa. Similar conclusions were drawn by Salmatonidis et al., (2019) during the exposure assessment of thermal spraying processes at industrial scale; where it was demonstrated that despite a fully operating LEV, when the enclosure of the spraying booth was degraded, fugitive emissions significantly impacted exposure in the worker area.

Thus, a combination of different factors (process parameters-feedstock, air flow rate, and enclosure) should be taken into account to improve the efficiency of mitigation measures under real-world conditions. Nevertheless, the most efficient measure is the ACH (coupling LEV with enclosure) as can be observed in Figure 7b, where the reduction of particle number with increase of ACH is evidenced.

(B) Laser ablation of ceramic tiles

Mitigation measure: the laser engraving set up was equipped with a 5.6 m^3 capturing hood, partially enclosed, with an integrated LEV system operating with a fixed extraction flowrate (2000 m^3 h^{-1}). The laser was located in an industrial building of 8000 m^3, naturally ventilated, where a previous screening (not shown) indicated that there were no additional significant NPs sources. Two experimental conditions were evaluated: with and without extraction.

Particle emissions: particles were generated during a repetitive batch process: each tile was ablated during approximately two minutes. Mean particle concentrations monitored in the exposure area reached $6*10^5/cm^3$ (maximum). Average concentrations (1-min) during the period with no extraction were $3.5*10^4/cm^3$ and mean particles size 175 nm (range 10-700 nm). When the LEV was fully operating the above values altered to $1.2*10^4/cm^3$ and 109 nm, respectively.

Effectiveness of the mitigation strategy: Figure 8 shows an evident reduction in particle number exposure concentrations, once the LEV system was activated, with an average efficiency of 65% over a 30-minute monitoring period (Table 3). The exposure reduction was lower than in case study A (with efficiency greater than 85%).The lower efficiency, compared to the thermal spraying, was probably due to worse enclosure in the laser ablation scenario and lower ventilation rate. This result shows the interdependence between extraction and source enclosure. Salmatonidis et al. (2018) demonstrated that during the laser ablation of ceramic tiles, lower extraction and no enclosure were sufficient to mitigate high particle emissions at laboratory-scale. Hence, since the scale of the scenarios might influence the effectiveness of control measures, the assessment in real industrial conditions becomes necessary.

3.4 Comparison with literature studies

Literature data regarding the efficiency of technological measures applied for occupational exposure reduction is relatively scarce, especially under real-world industrial conditions Therefore, a comparison of the obtained results was carried out with a number of studies focusing on the effectiveness of ventilation systems used for exposure reduction to manufactured nanomaterials (Table 5). The studies shown in Table 5 were carried out at laboratory scale, simulating real operating conditions. In the present work, the efficiency of LEV systems was strongly depending on the volume of air in working room and on ventilation rates. The achieved exposure reductions were in the range 65%-99% being lower than 99% reduction reported by Kim et al., (2007) and by Old and Methner (2008). The studies of Cena and Peters (2011) and of Tsai et al., (2010) reported efficiencies only qualitatively, as “good” or “low”. This review evidences that quantitative, experimental and real-world assessments of the efficiency of mitigation strategies is missing in the literature devoted to occupational exposure and NP safety research. Our results highlight the interdependence of different mitigation strategies (e.g., LEV and source enclosure), which are frequently implemented simultaneously in real-world industrial scenarios. Unless this kind of scenarios are characterized in detail and for an ample number of NP emission sources, the implementation of exposure modelling tools will be strongly hindered.

4. Conclusions

The effectiveness of different mitigation measures for NP exposure reduction was assessed in four industrial settings. The following conclusions can be drawn:

- The efficiency of common engineering control mitigation measures such as local exhaust ventilation (LEV) and source enclosure can vary significantly depending on the intensity of LEV (flowrate), the total volume of air in the exposure area, the type of enclosure (e.g. partial, total), and their combinations. Adequate LEV configurations may reduce exposure concentrations (in terms of particle number) by 65-85% and even reach 99% by combining higher flow rates and enhanced enclosure.

- Adequate maintenance operations and enhanced sealing were applied to an industrial kiln used for firing ceramic tiles. Source isolation based on improved sealing in the firing compartment reduced exposure concentrations by 52%. In addition, a new kiln operating with an enhanced sealed combustion hearth minimised NP release in the worker area down to 84% of the measured exposure concentrations. In this case study, however, particle number concentrations remained high after the implementation of the mitigation strategies (ca. 10^5 cm^{-3}). In spite of the fact that the presence of workers in the kiln zone is limited, additional measures would be required to improve workers' protection.
- The emissions from diesel engines significantly impact indoor the exposure to NPs. Substituting diesel with electric forklifts achieved a 92% reduction of particle number concentrations in breathing zone when the forklifts were in operation.

A review of the literature available evidenced the major need for real-world assessments of the efficiency of exposure mitigation strategies. One clear challenge identified is the interdependence of different strategies, which are frequently implemented simultaneously in industrial settings. The diversity of emission sources (stationary processes, moving vehicles, size of infrastructure, etc.) contribute to the complexity of this type of assessment. However, these data are necessary as input for exposure modelling and risk assessment tools.

Conflicts of interest

The authors declare no conflict of interest relating to the material presented in this article. Its contents, including any opinions and/ or conclusions expressed, are solely those of the authors.

Acknowledgements

The authors gratefully acknowledge the collaboration of the European Ceramics Centre in Limoges. This work was carried out in the framework of the CERASAFE project (www.cerasafe.eu), with the support of SIINN ERA-NET (project id: 16), and was funded by the Spanish MINECO (PCIN-2015-173-C02-01). Partial funding from the "Generalitat de Catalunya" (project number: AGAUR 2017 SGR41) is also acknowledged.

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Tables

Table 1. Particle emission scenarios and applied mitigation measures.

Case study	Activity-Source	Scale of facility	Mitigation measure
A	Atmospheric plasma spraying	Semi-industrial	Engineering controls: LEV & partial/full enclosure
B	Laser ablation	Industrial	Engineering controls: LEV & partial enclosure
C	Diesel forklifts	Industrial	Source substitution
D	Ceramic tile firing	Industrial	Source isolation: refurbishment& technology upgrade

Table 2. Instrumentation used for particle monitoring.

Instruments	Size range (nm)	Data recorded	Sampling locations
NanoScan-SMPS (TSI 3910)	10-420	Size resolved particle number concentration (#/cm^3)	Emission source Worker area
Condensation Particle Counter (CPC, TSI 3775)	4-3000	Total particle number concentration (#/cm^3)	Emission source Worker area
Diffusion Size Classifier miniature (DiSCmini, TESTO)	10-700	Particle number concentration (#/cm^3), mean diameter (D_p, nm)	Emission source Indoor background Outdoor background
Mini Laser Aerosol Spectrometer (Mini-LAS 11R, GRIMM)	250-32000	Size segregated mass concentration (µg/m^3)	Emission source Indoor background Outdoor background
Mini Wide Range Aerosol Spectrometer (Mini WRAS 1371, GRIMM)	10-32000	Size resolved particle number concentration (#/cm^3), Size segregated mass concentration (µg/m^3)	Indoor background Outdoor background

Table 3. Efficiency (reduction in particle number concentrations) of the mitigation strategies measured in operating conditions (NP: nanoparticle. LEV: local exhaust ventilation, air change per hour: ACH).

Mitigation measure	**NP source**	**Experimental conditions**	**Efficiency (%)**
Enhanced LEV with enclosure	Thermal spraying (A)	ACH=132 h^{-1}; door closed; exp. #2-#3	98.5-99.8%
Enhanced LEV with partial enclosure	Thermal spraying (A)	ACH=66 h^{-1}; door open; exp.#5	95.4%
LEV with partial enclosure	Thermal spraying (A)	ACH=33 h^{-1}; door open; exp.#4	85.6%
Enhanced LEV with partial enclosure	Thermal spraying (A)	ACH=66 h^{-1}; door close; exp.#1	99.3. %
LEV with partial enclosure	Laser ablation (B)	Extraction flowrate unavailable; partial enclosure	65.1%
Source substitution	Diesel forklifts (C)	Only for driving periods	91.5%
Source substitution	Diesel forklifts (C)	Average of driving and stationary periods, 1 hours	48.7%
Source isolation	Ceramic tile firing (D)	Enhanced sealing of the kiln	51.6%
Source isolation	Ceramic tile firing (D)	Optimal sealing of kiln and superior refractory condition	84.4%

Table 4. Mean particle number concentrations inside the spraying room and in the exposure area (worker room), and experimental details for each of the experimental runs (scenario A, LEV: local exhaust ventilation, air change per hour: ACH).

Experiment parameters					**Particle number concentration (cm^{-3})**	
Run	**Feedstock**	**LEV flowrate (m^3/h)**	**ACH (h^{-1})**	**Interior door**	**Spraying room**	**Worker room**
#1	NiCrAlY	12000	66	closed	9.2×10^5	6.9×10^3
#2	NiCrAlY	24000	132	closed	1.5×10^6	3.3×10^3
#3	ZrO_2+4mol%Y_2O_3	24000	132	closed	6.6×10^5	5.2×10^3
#4	$Zn(NO_3)_2 \cdot 6H_2O$	12000	33	opened	3.7×10^5	5.4×10^4

#5	$Zn(NO_3)_2 \cdot 6H_2O$	24000	66	opened	5.0 x 10^5	2.3 x 10^4

Table 5. Review of literature studies on the efficiency of ventilation systems for exposure reduction when dealing with manufactured nanomaterials (MNMs). The multiwalled carbon nanotubes (MWCNTs) had lengths between 1-20 nm and the D_p corresponds to their outer diameter.

Mitigation measure	**Configuration**	**NP type**	**Size (nm)**	**Efficiency**	**Reference**
Movable LEV system	-	Ag, Mn, Co	300	>99%	Old and Mehner, 2008.
Constant velocity hood	Constant hood face velocity = 0.5 m/s	Al_2O_3	200	Good performance	Tsai et al, 2010.
Constant flow hood	Constant airflow, hood face velocity varies inversely with height of sash opening	Al_2O_3	200	Low performance	Tsai et al, 2010.
Biological safety cabin		MWCNTs	D_p: 10-50	Good performance	Cena and Peters, 2011
Filters used in fume hoods (HEPA)	-	Ag	10	>99.99%	Kim et al, 2007

Figures and Captions

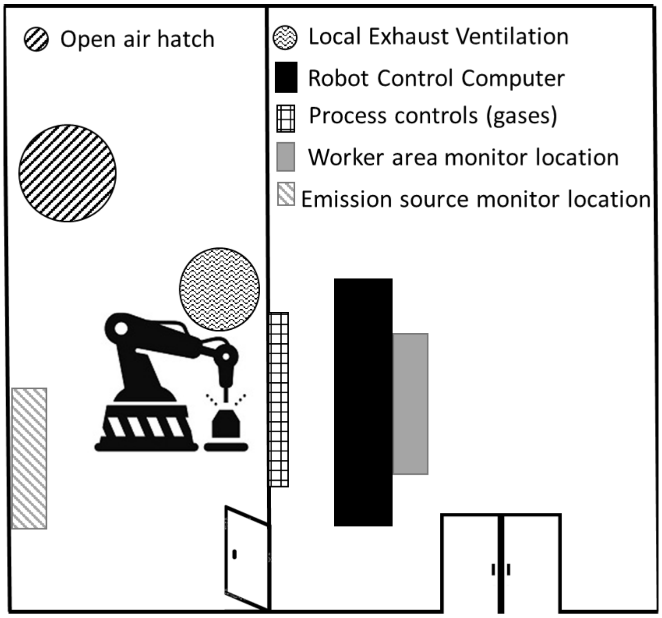

Figure 1. Schematic illustration of the APS facility (scenario A), spraying room (left) and worker room (right).

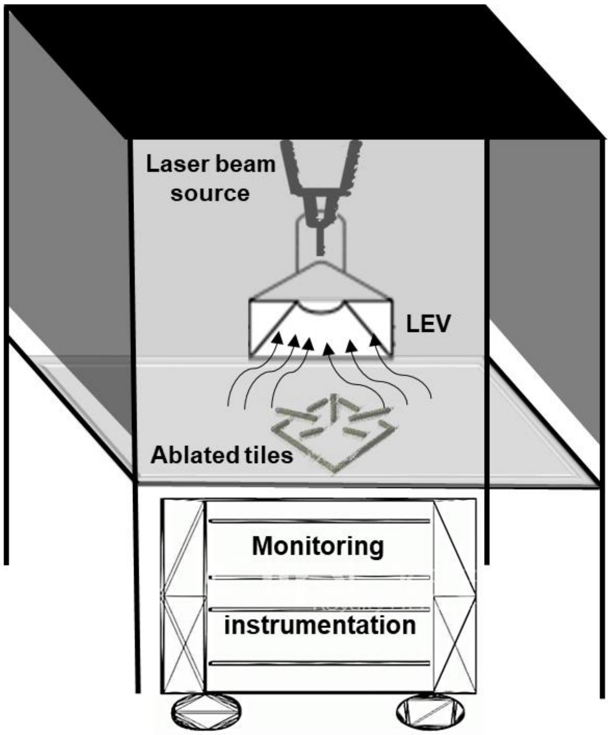

Figure 2. Schematic illustration of the measurement set-up during laser ablation of tiles (scenario B).

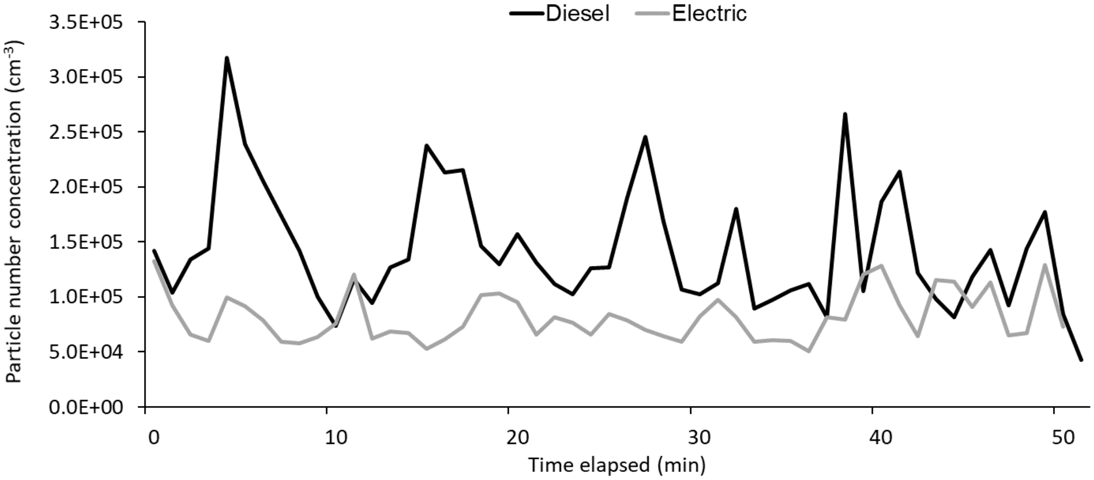

Figure 3. Particle number concentrations monitored in the worker breathing zone (scenario C), at operation using a diesel (black) and an electric (grey) forklift.

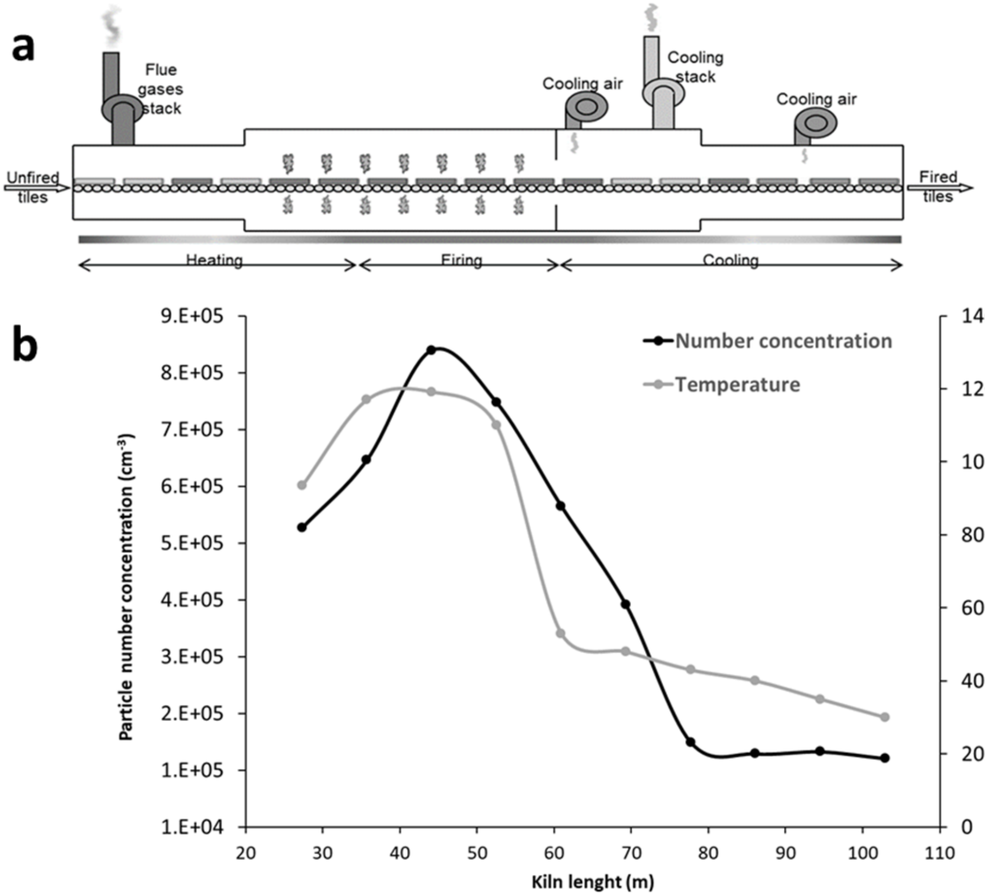

Figure 4. Schematic illustration a roller hearth kiln (scenario D) with the corresponding nanoparticle release (a) and temperature along the kiln (b).

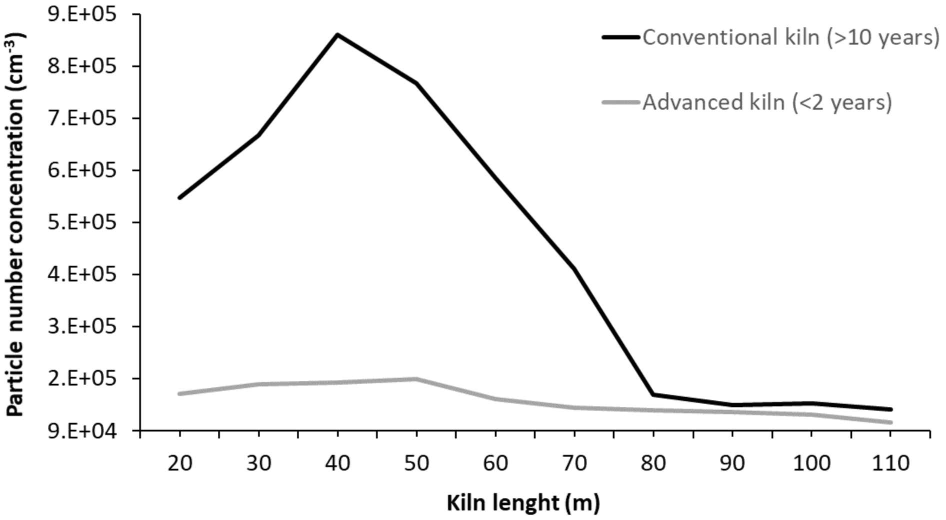

Figure 5. Emissions of particle in terms of number concentration along two kilns (scenario D): conventional (black curve) and advanced (grey curve). The peak at 45 m corresponds to the highest temperature zone (firing).

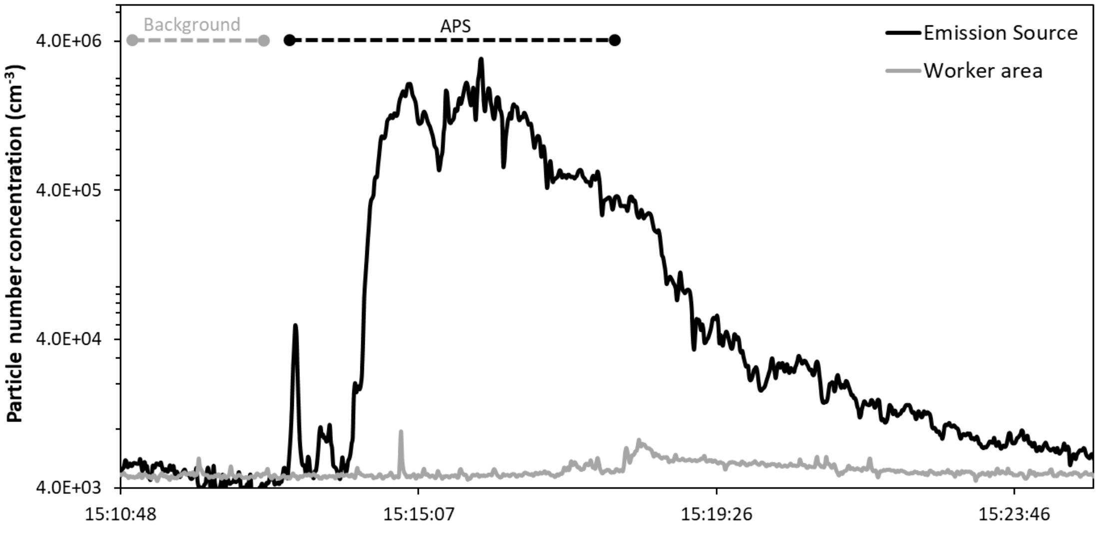

Figure 6. Particle number concentrations (10-700nm with DiscMini) for the experiment #2 inside the spraying room (emission source), and in the worker area (scenario A).

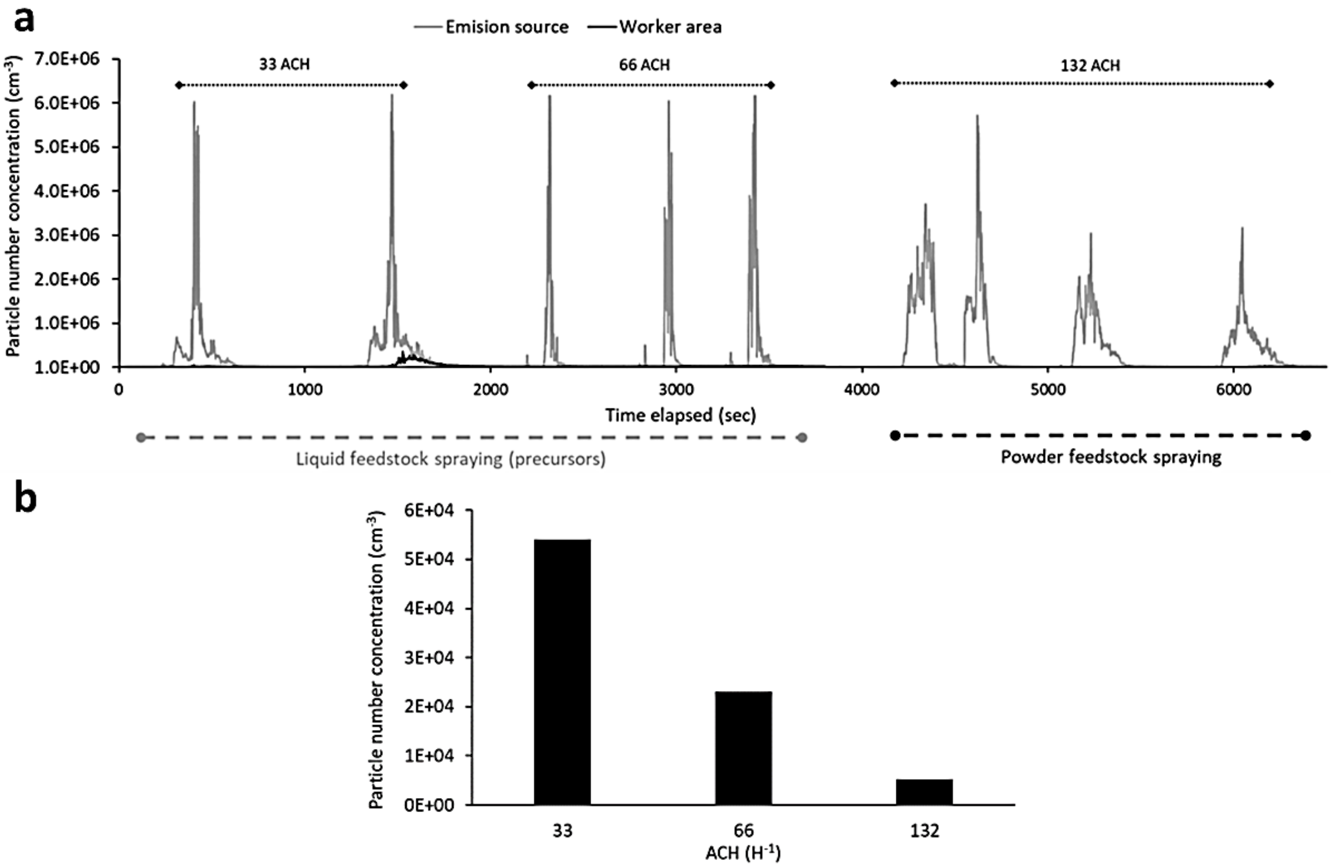

Figure 7. (a) Particle number concentrations (10-700nm with DiscMini) for the experiments #6, #7 and #5 from left to right, (b) number concentration in the worker area for different ACH values (scenario A).

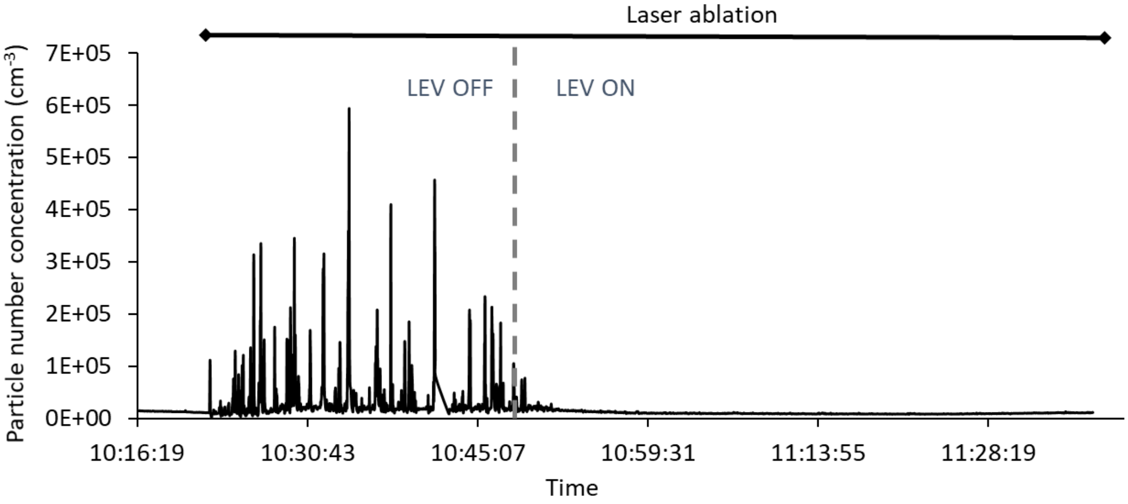

Figure 8. Particle number concentrations monitored with and without local exhaust ventilation (LEV; scenario A).

Effectiveness of nanoparticle exposure mitigation measures in industrial settings

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Supplementary material

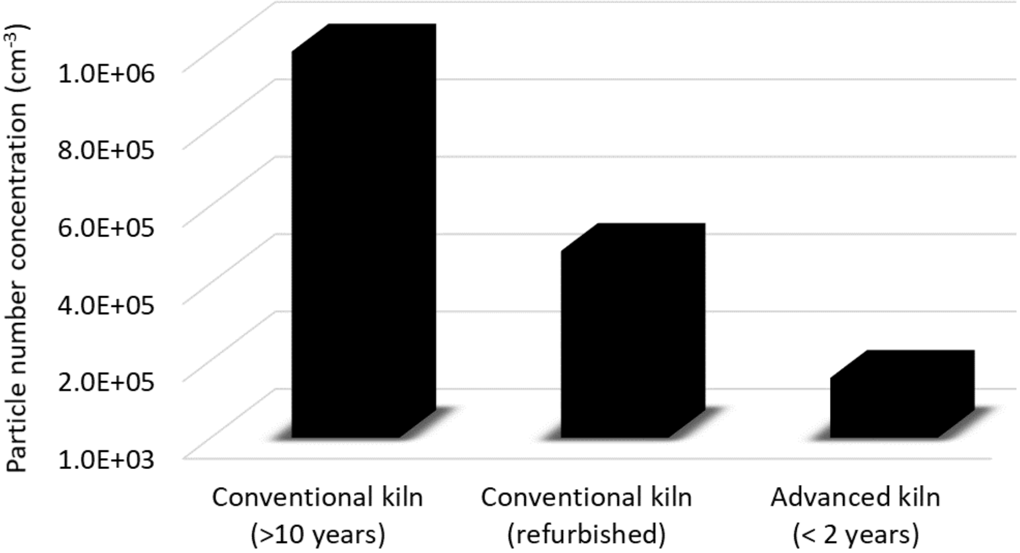

Figure S1. Evolution of emissions in terms of mean particle number concentration in three kilns having different isolations, refractory conditions and number of service years. The decreasing trend of particle release with improved isolation of the firing zone and refractory condition, can be observed.

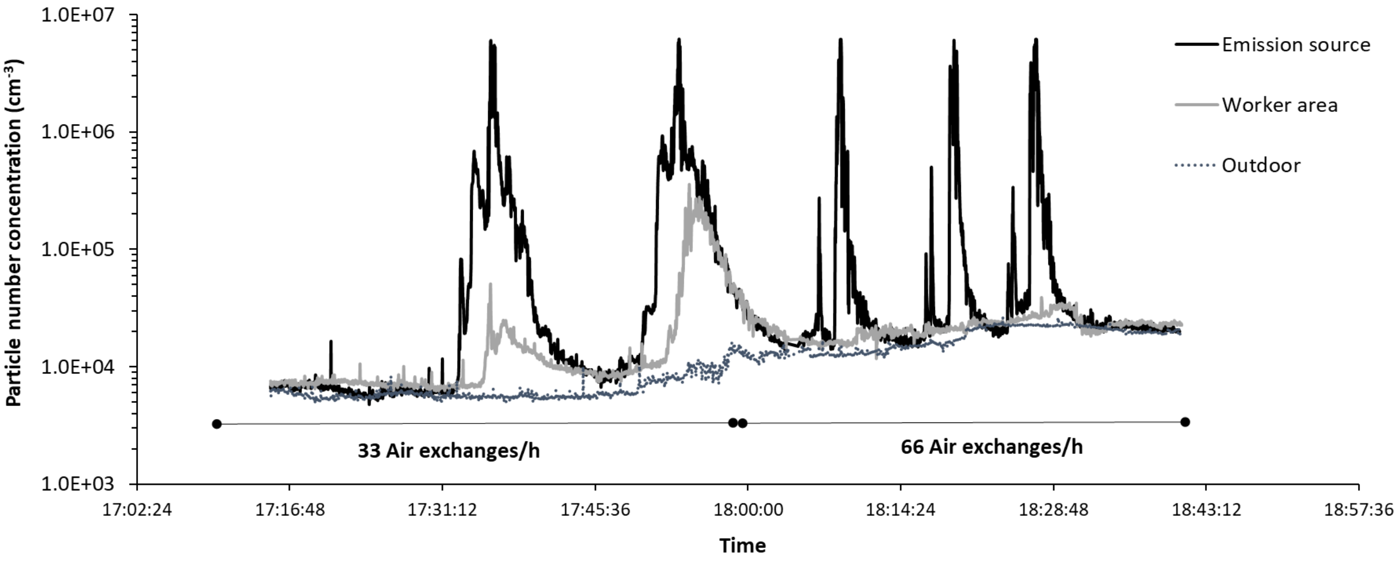

Figure S2. Emissions expressed in particle number concentrations monitored simultaneously during APS processing at the emission source (black curve), in the worker area (gray curve) and outdoor (doted curve). The experiments #4 and #5 have different LEV flow rates (12000/24000 m^3/h) but the same enclosure conditions (door open) under the spraying of the same feedstock (scenario A).