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Original article

Measurement of Airborne Particles and Volatile Organic Compounds Produced During the Heat Treatment Process in Manufacturing Welding Materials

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ABSTRACT

Background: There is little information about the airborne hazardous agents released during the heat treatment when manufacturing a welding material. This study aimed to evaluate the airborne hazardous agents generated at welding material manufacturing sites through area sampling.
Methods: concentration of airborne particles was measured using a scanning mobility particle sizer and optical particle sizer. Total suspended particles (TSP) and respirable dust samples were collected on polyvinyl chloride filters and weighed to measure the mass concentrations. Volatile organic compounds and heavy metals were analyzed using a gas chromatography mass spectrometer and inductively coupled plasma mass spectrometer, respectively.
Results: The average mass concentration of TSP was 683.1 ± 677.4 μg/m^3, with respirable dust accounting for 38.6% of the TSP. The average concentration of the airborne particles less than 10 μm in diameter was 11.2–22.8 × 10^4 particles/cm^3, and the average number of the particles with a diameter of 10–100 nm was approximately 78–86% of the total measured particles (<10 μm). In the case of volatile organic compounds, the heat treatment process concentration was significantly higher ($p < 0.05$) during combustion than during cooling. The airborne heavy metal concentrations differed depending on the materials used for heat treatment. The content of heavy metals in the airborne particles was approximately 32.6%.
Conclusions: Nanoparticle exposure increased as the number of particles in the air around the heat treatment process increases, and the ratio of heavy metals in dust generated after the heat treatment process is high, which may adversely affect workers' health.

1. Introduction

In developed countries, the proportion of traditional secondary manufacturing industries is decreasing compared to that of high-tech industries, such as tertiary industries and electronics companies [1]. However, industries that form the foundation for manufacturing industries, such as welding and heat treatment, are important in all countries. The Republic of Korea proposed the Root Industry Promotion and Advancement Act (Root Industry Act) with six underlying industries (heat treatment, molding, welding, plastic working, casting, and surface treatment) as root industries. Amidst rapid growth, the potentially hazardous agents produced by these industries have not been assessed, mainly, because the relevant policy was proposed by the Ministry of Trade, Industry and Energy Act, which focuses more on the promotion of the industry than the industrial health and safety of workers. To ensure that workers can work safely and the sector can grow sustainably, the occurrence of and exposure to hazardous agents need to be assessed and minimized.

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Heat treatment refers to the process of heating and cooling a material to certain temperatures to improve its specific properties or metal structure. It is a basic process used in most industrial settings, such as in wood and surface treatment, small parts manufacturing, and large equipment manufacturing [2–5]. Heat treatment can be widely classified into four types: cascading temperature rising, constant temperature, continuous cooling, and case hardening. Typical heat treatment consists of four stages: quenching, tempering, normalizing, and annealing. Quenching increases the hardness and strength of a material, tempering prevents the risk of breakage and stabilizes the object tissue by granting toughness to materials with increased hardness after the quenching process. Normalizing makes the material into a standard tissue and reduces deformation after quenching, and annealing softens materials.

According to previous studies and national on-site evaluation data, various volatile organic compounds (VOCs) and heavy metals are generated during the heat treatment process in the workplace along with dust from the ambient air [6]. VOCs are mainly generated during the burning process before heat treatment. During the heat treatment process, airborne particles are formed in the process of changing the structure and arrangement of the atoms in the metal due to heating and cooling [7]. Particles with a particle size of 7 to 30 nm occur between 350 and 500°C [8,9]. As the temperature of the heat treatment increases, the number of particles with a larger diameter increased and the surface of the particles became rougher [10]. During softening in the heat treatment process, and in this process, metal particles may occur, with sodium (Na) and strontium (Sr) components detected by the components used inside the metal [11,12]. Other heavy metals are generated during the heat treatment process. Prolonged exposure to heavy metals in the air, such as cadmium (Cd), cobalt (Co), and lead (Pb), has been proven to cause deoxyribonucleic acid damage and genotoxic effects [13–15]. In addition, Cd and Pb have a significant effect on causing cough and lead on causing asthma [16,17]. Some diseases from heavy metal exposure show rapid symptoms, but there are cases where workers who were exposed to heavy metals suffered from health problems after retirement.

Extensive research has been conducted on industrial development worldwide; however, this generally focused on technological development and the use of heat treatment in manufacturing, rather than on the health effects of hazardous chemicals on industry personnel. Airborne particles containing heavy metals, VOCs, and heat stress are known to be hazardous. While the impacts of heat stress have been extensively studied, the impacts of exposures to airborne particles and VOCs have not [18,19]. Therefore, this study aimed to evaluate the effect on workers of exposure to airborne hazardous agents in the workplaces while conducting heat treatment.

2. Materials and methods

2.1. Sampling sites

Measurements were conducted at a workplace that conducts heat treatment for manufacturing flux-cored wires (FCWs), a welding material in the Korean industrial complex. The concentration of airborne hazardous agents was measured for three heat treatment processes at least 3 h. The heat treatment process in the field had a protocol to repeat the same task for 24 h at a fixed position. As a result, the workers regularly stayed within 1 m from the treatment mill for more than one hour while working. Sampling was performed at a height of 1.5 m, where the worker's breathing zone is located. The top view of the heat treatment process is shown in Fig. 1 and a schematic representation is shown in Fig. S2.

Table 1 shows the main characteristics of the heat treatment process for three sampling sites. As the stainless steel (SS) and mild steel (MS) heat treatment processes include a liquid cooling step for surface softening and impurity removal at the solvent material's outlet, hazardous agents in the surrounding air were assessed. Both SS and MS heat treatment are bright annealing processes, and the material introduced into the process undergoes surface softening at

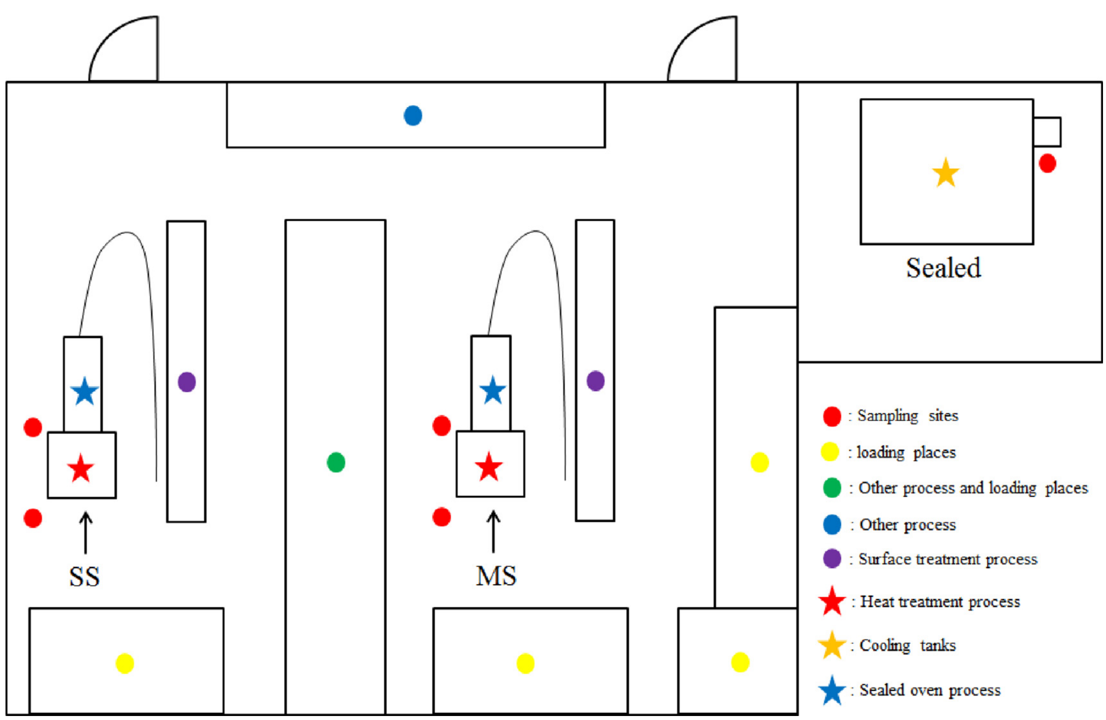

Fig. 1. The top view of the heat treatment process and sampling sites: the stainless steel and mild steel heat treatment processes and the Sealed oven process.

Table 1
Heat treatment conditions for three sampling sites (flux-cored wires, FCW)

	Temperature	Retention time*	Status	Material	Type†
Stainless steel process	1050°C	7–15 s	Open	FCW (stainless steel)	O
Mild stessl process	1050°C	7–15 s	Open	FCW (mild steel)	O
Sealed process	450°C	8 h	Sealed	FCW (Mix)	X

Abbreviations: FCW, flux-cored wire.
* Retention time: indicates how long the wire stays in the heat treatment process.
† Type: divided into inlet and outlet.

1050°C for 7–15 s. However, the processes differ in the type of material processed: SS heat treatment processes FCW made of stainless steel, while MS heat treatment processes FCW made of MS. Owing to the nature of the sealed site procedure, which involves heat treatment inside a facility, measurements were made in the vicinity of the control bar where employees monitored and managed the facility. The sealed site method involved baking (final heat treatment process) at 450°C for 8 h in a sealed facility.

2.2. Sampling and analysis

2.2.1. Gravimetric analysis

For measuring total suspended particle (TSP) and respirable dust (RD) concentrations, samplings were performed at a height of 1.5 m using an air sampling pump (Gillian, Sensidyne, USA). The NIOSH NMAM 0500 was selected to sample the TSP for applying a respiratory rate of 2 min/L using a polyvinyl chloride filter (37 mm, pore size 5 μm, SKC, USA) on a support pad and 3 piece-cassette (Whatman Grade QM-A, 37 mm; Whatman, Midstone, UK). The RD samples were collected using a polyvinyl chloride filter as TSP but the flow rate was 2.5 L/min by attaching a respirable cyclone. Before and after the sampling, filters were quantified according to NMAMs 0500 & 0600 and weighed using an electronic scale (Mettler Toledo, AB204-S, Switzerland) with an accuracy up to 1 μg [20,21]. In addition, each pump's flow rate was calibrated using air flow calibration (Drycal, Mesa Labs Bios Defender 510-H, Primary Flow Meter, USA) before and after the sampling. The mass concentration was obtained by dividing the measured weight by the sampling volume, which can be calculated from the sampling flow rate and time.

2.2.2. Real-time monitoring of airborne particles

Real-time monitoring was carried out using an optical particle sizer (OPS, Model 3330, TSI Inc., USA) for a size range of 0.3–10 μm. A scanning mobility particle sizer (SMPS, Model 3910, TSI Inc., USA), which can assess a size range of 10–420 nm, was used to evaluate the number concentration of the fine airborne particles, including the nano materials in the air at each site.

2.2.3. VOCs

VOCs samples were collected using a low air volume pump (Gillian) at a flow rate of 0.2 L/min using a charcoal tube (ANASORB CSC. Coconut Shell Charcoal. Cat No. 226-01, SKC Inc., USA) and moved using an ice box (−5 to −10°C). The content of the charcoal tubes was desorbed using 1 mL carbon disulfide (CS_2), and solid particles were filtered using an Advantec disposable membrane filter (13JP050AN. Lot No. 907111BD, pore size 0.22 μm). After this pretreatment, the samples were analyzed by a gas chromatography mass spectrometer (GC-MS 7890A, Agilent) using the DB-5MS UI Column using the liquid injection method.

2.2.4. Metals

The polyvinyl chloride filters after weighing for a gravimetric analysis were analyzed according to the NIOSH NMAM 7304 method. Each filter was put into a polytetrafluoroethylene vessel and 5 mL of 70% nitric acid was injected for acidic digestion using a Mars System (CEM, model no. 910900, Mathews. NC, USA) [22]. Then, the suspension was diluted to a volume of 35 mL with distilled water. For each sample, a 1 mL solution was diluted to a total volume of 10 mL at a ratio of 1:9 with 5% distilled water. The samples were then analyzed using an inductively coupled plasma mass spectrometer (Optima 3000, Perkin-Elmer, USA).

2.3. Statistical analysis

Statistical analyses were conducted using the R 3.6.3 program for data obtained through measurement and analysis. Analysis of variance was conducted to verify the difference between the descriptive statistics of the data and the measured values at each measurement location. In the case of the Analysis of variancea-nalysis, the Kruskal–Wallis test was used for a statistical analysis after testing the normality of the data. The Bonferroni method was used for a *post-hoc* analysis. Statistical significance was set at $p < 0.05$.

2.4. Comparison with exposure standards

The air concentrations of hazardous materials, such as airborne particles, heavy metals, and VOCs, were compared with the Occupational Exposure Limits (OELs) designated by the Ministry of Employment and Labor in the Republic of Korea.

Table 2
The condition of the process status for three sites

Category		*Sampling time/sample (min)	N (no. of samples)	Temperature at the measurement site (°C)	Relative humidity (%)	Ventilation type
Stainless steel	Inlet	243 ± 28.7	4	27.0 ± 1.6	14.2 ± 5.7	Natural
	Outlet	205 ± 27.6	4	23.7 ± 0.9	17.8 ± 5.2	Natural
Mild steel	Inlet	247 ± 28.5	4	28.2 ± 1.2	14.0 ± 5.8	Natural
	Outlet	193 ± 34.1	4	23.8 ± 2.1	21.1 ± 6.9	Natural
Sealed	—	233 ± 27.4	8	33.9 ± 1.3	12.3 ± 3.6	Natural
Outside	—	469 ± 42.9	4	11.0 ± 5.0	46.4 ± 14.0	—

* Sampling time: average sampling time per each sampling.

3. Results

3.1. Temperature and humidity at sampling sites

Table 2 summarizes the conditions for the measurement environment of each sampling site. In the heat treatment process for both SS and MS, combustion was performed before cooling. Evaluation was conducted at the Inlet and Outlet. At the Inlet, near the combustion location, the temperature was high and humidity was low. In contrast, the Outlet was cooled by the liquid, causing low temperature and high humidity. Overall, high average temperature and low humidity were observed during the Sealed process.

3.2. Gravimetric analysis of airborne particles

Data on airborne particles, measured as TSP and RD, are summarized in Table 3. The average mass concentration of TSP was 683.1 $\pm$ 677.4 $\mu g/m^3$ and the average concentration of RD was 263.4 $\pm$ 368.5 $\mu g/m^3$, accounting for 38.6 % of TSP. The highest TSP concentration was observed at the SS Outlet (2026.1 $\pm$ 470.3 $\mu g/m^3$) and the lowest was Outside (205.3 $\pm$ 57.6 $\mu g/m^3$), and the concentrations at these sites were significantly different from those at other sites ($p < 0.05$). The highest RD concentration was observed at the SS Outlet (1000.3 $\pm$ 298.1 $\mu g/m^3$) and the lowest at the MS Outlet (25.4 $\pm$ 22.21 $\mu g/m^3$), and these values were statistically and significantly different from those at other sampling sites ($p < 0.05$).

3.3. Number concentration of airborne particles

Fig. 2 shows the average number concentration distribution (dN/dlogDp) during the measurement period for different sizes of airborne particles at each sampling site. Dp means the size of the particles and N means the number concentration of the particles. Most of the airborne particles collected at each sampling site had a size of 100 nm or less, statistically higher than that Outside (control group; $p < 0.05$). In addition, for bright annealing SS, the Outlet region, close to where the quenching process occurs, had a higher number concentration than the Inlet region, where combustion occurs. Comparatively, bright annealing MS showed the opposite results.

3.4. VOCs

Table S1 shows the VOC measurement and analysis results for each sampling site. Nine VOCs were identifiable (bromodichloromethane, naphthalene, ethylbenzene, p-xylene, m-xylene, styrene, o-xylene, benzene, and toluene), with other materials not reaching the limit of detection. The VOC concentrations are presented in Table S1 and Fig. 3. In the case of the SS and MS Inlet, the concentration of the VOC was higher than that at Outlet sites. In particular, p-xylene, naphthalene, and toluene had higher air concentrations than other materials. Toluene had a concentration of approximately 1 $\mu g/m^3$ or more in areas excluding the SS Outlet and Sealed site. The concentration of airborne VOCs near the combustion process was high. In the case of the Sealed site, the concentration of the VOCs was about 40.1% and 48.5% of that at the SS and MS Inlets, respectively.

3.5. Heavy metals

Table S2 shows the measurement and analysis results of heavy metals. Ten metals [Chromium (Cr), Manganese (Mn), Iron (Fe), Aluminum (Al), Barium (Ba), Strontium (Sr), Sodium (Na), Potassium (K), Calcium (Ca), and Magnesium (Mg)] were observed. The concentrations of Fe, Na, Ca, and Mg, were relatively higher in the air compared with those of other metals. In addition, it was confirmed that the concentration of Fe (113.0 $\pm$ 148.2 $\mu g/m^3$) and Al (184.3 $\pm$ 192.6 $\mu g/m^3$) at the SS Inlet and that of Mg (168.5 $\pm$ 124.4 $\mu g/m^3$) at the MS Outlet was higher compared with those at other sampling sites. Moreover, most of the heavy metal materials had high airborne concentrations near the SS process. Comparatively, the Sealed site showed low airborne heavy metal concentrations.

As shown in Fig. 4, the concentration of total airborne heavy metals was the highest at the SS Inlet. Similarly, the concentrations of heavy metals were higher near the SS process than those near the MS process. In the case of MS, the Outlet concentration of airborne heavy metals was higher than the Inlet. The Sealed site showed similar results compared with the Outside. Most of the sites showed the highest percentage of Mg.

3.6. Comparison with exposure limit

The results compared with the OELs are shown in Table S3. Compared with OELs, most metals did not exceed the standard levels except for Sr. The concentration of strontium at all sites where measurements were conducted at the standard levels. The concentration near the heat treatment process was substantially greater. The concentration of heavy metals in the Outside was also measured to be high.

4. Discussion

Airborne particles generated during heat treatment are classified as particulates not otherwise specified. The Republic of Korea Domestic Occupational Safety and Health Act sets a limit for the concentration of particulates not otherwise specified of up to 10 mg/m^3. The concentration of the airborne particles generated in the heat treatment process in this study was well below this limit. However, heavy metals accounted for approximately 32.6% of the particles in the air. Additionally, the heat treatment process runs around the clock at the location and relies on natural ventilation. Workers were thus concerned about the health impacts of prolonged exposure to airborne hazardous airborne particles in this workplace, such as increased rates of cancer and respiratory disorders [23].

The integrated sampling method only revealed the total weight of the airborne particles generated in the process; however, the risk

Table 3
The average mass concentration of airborne TSP and RD by each site ($\mu g/m^3$)

Average	Stainless steel site		Mild steel site		Sealed site	Outside
	Inlet	Outlet	Inlet	Outlet		
TSP	700.9 $\pm$ 336.0	2026.1 $\pm$ 470.3	353.0 $\pm$ 97.7	404.6 $\pm$ 128.7	408.8 $\pm$ 375.1	205.3 $\pm$ 57.6
RD	165.6 $\pm$ 167.6	1000.3 $\pm$ 298.1	216.9 $\pm$ 261.0	25.4 $\pm$ 22.2	100.9 $\pm$ 129.7	38.0 $\pm$ 43.1

Described as follow: arithmetic mean $\pm$ standard deviation.
Abbreviations: RD, respirable dust; TSP, total suspended particle.

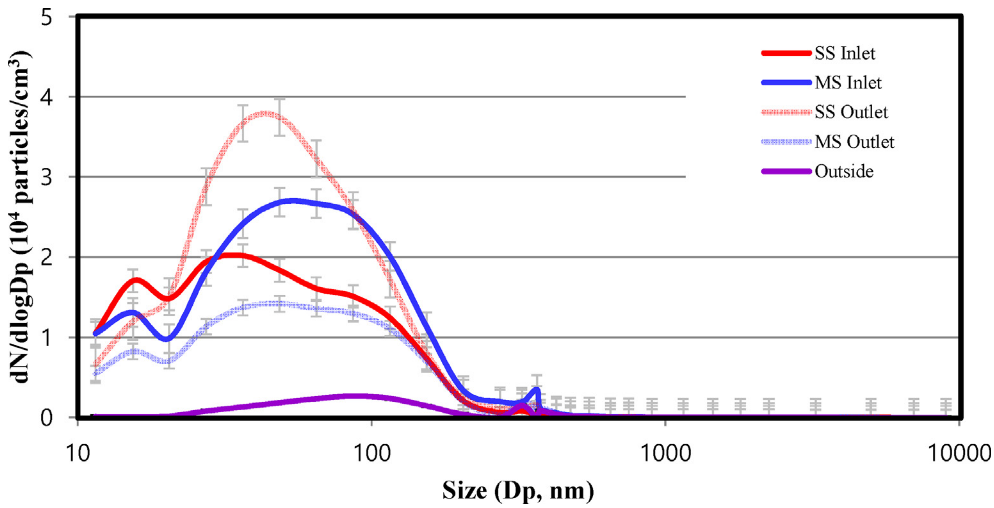

Fig. 2. The average number size distribution of airborne particles during the sampling period by size in the SS and MS processes. MS, mild steel; SS, stainless steel.

associated with the process remained obscure. To overcome this problem, real-time monitoring devices, usually not used to assess traditional industrial health, were utilized. These results revealed a variation in the number concentration of the particles by size. The average number concentration of the airborne particles with a size of 10–300 nm measured by SMPS was 4.3–87.4 times higher than the average number concentration for the particles with a size of 300–10,000 nm measured by OPS. Less than 50% of the nanomaterial particles with a size of 10 nm are deposited in the lung alveoli, compared to less than 20% for the particles with a size of 100 nm or more. Health hazards associated with nanoparticles have been extensively reported [24]. As shown in Table 4, for the Inlets and Outlets evaluated in this study, the airborne particles corresponding to the sizes of 10–100 nm accounted for 77–86% of all particles, significantly higher than that at workplaces investigated in previous studies [25–28]. In addition, the contribution of nanoparticles (100 nm) in airborne particles during heat treatment was higher than in other workplaces, indicating that there are many fine particles present even when the mass concentration of airborne particles is low. Particles with a larger diameter are generated as the temperature of the heat treatment process increased, but nanoparticles are mainly distributed by the particle size. Nanoparticles were mainly generated in the heat treatment Inlet and Outlet process. The working environment, which is difficult to control these particles, can increase the nanoparticle exposure of workers. Therefore, a management to control and prevent exposure to airborne particles through isolation, sealing, and installation of local exhaust devices is important at work sites. Moreover, the provision of personal protective equipment is suggested [29].

Among VOCs, toluene had a high concentration at each site. Since toluene can cause neurological illnesses, workers who spend long hours close to the site were worried about the health impacts, even though the concentrations did not reach the OELs set by the Ministry of Employment and Labor in the Republic of Korea [30]. The heat treatment was performed at 1000°C, which was sufficient to change the components of the metal. The VOCs generated due to the heating metal components used for the desired properties and functions may be directly exposed and be generated through cracks in the metal generated during heating. In addition, VOCs may occur in the heating part before heat treatment and in the subsequent cooling process, and the fact that both are in an open state increases the possibility of worker's exposure to VOCs. Additionally, benzene exposure has been linked to cardiovascular disease and cancer; thus, its management may be required [31,32]. The evaluation showed that VOCs did not exceed the exposure standard even though combustion was performed openly in the workplace before the heat treatment was carried out, and ventilation was carried out

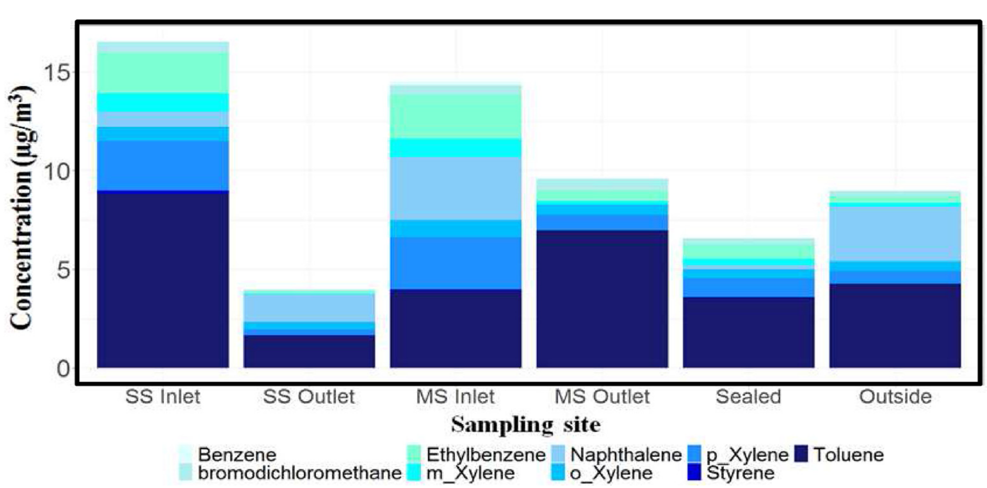

Fig. 3. The concentration of VOCs by the sampling site. VOCs, volatile organic compounds.

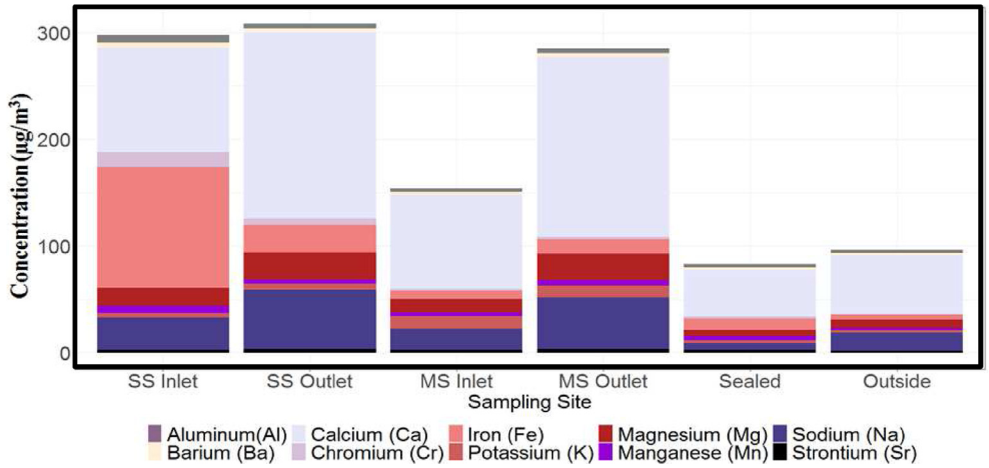

Fig. 4. The concentration of heavy metals by the sampling site.

Table 4
The average number concentration of airborne particles by size

Site		Size range(nm)					Nano fraction (%)
		10–100 (10^3 particles/cm^3)	100–300 (10^3 particles/cm^3)	300–1000 (10^3 particles/cm^3)	1000–10,000 (particles/cm^3)	Total (10^4 particles/cm^3)	
SS	Inlet	131.5 ± 3.1	23.2 ± 5.0	0.9 ± 0.1	69.7 ± 6.6	15.5 ± 0.8	84.7
	Outlet	154.6 ± 7.3	41.3 ± 7.4	4.6 ± 1.3	51.4 ± 5.3	19.7 ± 1.0	78.4
MS	Inlet	194.6 ± 11.7	31.1 ± 6.5	2.3 ± 0.6	45.1 ± 4.5	22.6 ± 1.3	85.9
	Outlet	86.5 ± 3.4	22.6 ± 4.3	2.3 ± 0.4	62.8 ± 6.6	11.0 ± 0.5	78.4
Outside	—	9.3 ± 1.1	5.0 ± 0.9	0.8 ± 0.1	43.4 ± 4.9	1.5 ± 0.08	61.5

Abbreviations: SS, stainless steel; MS, mild steel.

naturally. It is necessary to establish data on VOCs in the air during heat treatment at other work sites because despite the global spread and utilization of this process, there have been fewer prior studies and epidemiological investigations on the associated hazardous agents.

Sr showed the highest airborne concentration among heavy metals. Apart from accumulating in the bones, Sr primarily enters the human body through ingestion, respiration, and the skin. Moreover, excessive exposure to strontium can result in kidney failure [33]. Cr (VI) exposure can induce oxidative stress in the body and cause Fe metabolic disorders, kidney damage, and cancer [34–36]. Fig. 4 shows that the concentration of Fe at the SS Inlet and Outlet was particularly high. The SS that is employed in the SS process is occasionally cut off as it is being inserted into the SS Inlet and a bundle of iron is loaded, leading to this high concentration. In addition, the concentration of airborne heavy metals was significantly low at the Sealed site, which was attributed largely to the characteristics of the Sealed process.

Heavy metals showed the opposite trend to that of VOCs at each site (Table S3). It is challenging to conclude that the airborne heavy metals produced during the heat treatment process did not have an influence after dividing the generated chemicals by each regulatory level and summing them up because the total for all processes exceeded 1. Some high toxicity materials produced by the heat treatment procedure may also not have been analyzed. According to estimates, the high value in the Outside air measurement was caused by the fact that tests were taken inside an industrial complex and that the steel was frequently transported nearby.

The RD/TSP and heavy metals (HM)/TSP ratios among each site are shown in Table 5. The RD/TSP ratio, that is the ratio of airborne particles that could be inhaled and deposited to the human respiratory area, showed no statistically significant difference between the Inlet and Outlet. However, the value of HM/TSP at the Outlet (0.697) was higher ($p < 0.2$) than that at the Inlet. Therefore, it could be more harmful to be exposed to airborne particles around the Outlet. Although the RD/TSP ratio between the entire heat treatment process and the baking (Sealed site) was similar, the HM/TSP value implied that the heat treatment process (0.581) was statistically significant ($p < 0.06$), that is, the heat treatment process could be more harmful. Finally, the 100 nm/10 μm value, the proportion of the nano materials in the particles exposed to the respiratory area, was significantly different at the Inlet and Outlet. As the number of airborne particles that can be exposed to the respiratory area increases, the tendency to increase nanoparticle exposure suggests a statistically strong association (Table 6).

Table 5
The ratio of each parameter and *p*-value of each case

	Ratio of each parameter	*p*
RD/TSP		
Inlet	0.380	—
Outlet	0.290	
Entire heat treatment process	0.335	—
Baking (sealed site)	0.383	
Heavy metals/TSP		
Inlet	0.465	0.20
Outlet	0.697	
Entire heat treatment process	0.581	0.06
Baking (sealed site)	0.317	

Abbreviation: RD, respirable dust; TSP, total suspended particle.

Table 6
The Pearson's correlation coefficient and ratio of each parameter of each case

	Pearson's r	Ratio of each parameter
100 nm/10 μm		
Inlet	0.997	0.334
Outlet	0.996	0.318
Entire heat treatment process	0.996	0.326

The study had limitations owing to the limited sample size and measurement duration, and in the case of the evaluated procedure, the limited workload per day and hour makes it challenging to represent the measured values of the heat treatment process. Additionally, it was difficult to choose the materials to be analyzed because the composition information for products and additives in the process was restricted as the company's trade secrets, and it was only possible to guess where VOCs and heavy metal substances could be identified. It was challenging to determine the precise exposure of workers by collecting local samples, despite the fact that identical sampling was performed for 24 h owing to the stationary sampling positions.

In summary, the VOC concentration at the Inlet was significantly higher than that at the Outlet. Compared to the Outlet of other parts at the Sealed site, 40–49% of VOCs were detected, making it difficult to say that the Sealed process could effectively shield against gaseous substances. In the case of heavy metals, the concentration of heavy metals discharged to the Outside could be reduced if there was a barrier. Even though the RD/TSP ratio of the Inlet and Outlet was similar, the HM/TSP ratio showed a moderate statistical difference ($p < 0.2$). Particularly, the HM/TSP ratio was higher around the Outlet (0.697) than the Inlet part (0.465) during the heat treatment process. The HM/TSP ratio observed around during heat treatment was significantly higher than that observed during baking (Sealed site) ($p < 0.06$). The health hazards to workers during the heat treatment process should be assessed in a follow-up study applying individual sample measurements.

Conflicts of interest

All authors have no conflicts of interest to declare.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.shaw.2023.03.005.

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