

Particle Size Distributions of Particulate Emissions from the Ferroalloy Industry Evaluated by Electrical Low Pressure Impactor (ELPI)

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The present article presents a comprehensive evaluation of the potential use of an Electrical Low Pressure Impactor (ELPI) in the ferroalloy industry with respect to indoor air quality and fugitive emission control. The ELPI was used to assess particulate emission properties, particularly of the fine particles ($D_p \leq 1 \mu\text{m}$), which in turn may enable more satisfactory risk assessments for the indoor working conditions in the ferroalloy industry. An ELPI has been applied to characterize the fume in two different ferroalloy plants, one producing silicomanganese (SiMn) alloys and one producing ferrosilicon (FeSi) alloys. The impactor classifies the particles according to their aerodynamic diameter and gives real-time particle size distributions (PSD). The PSD based on both number and mass concentrations are shown and compared. Collected particles have also been analyzed by transmission and scanning electron microscopy with energy dispersive spectroscopy. From the ELPI classification, particle size distributions in the range 7 nm – 10 μm have been established for industrial SiMn and FeSi fumes. Due to the extremely low masses of the ultrafine particles, the number and mass concentration PSD are significantly different. The average aerodynamic diameters for the FeSi and the SiMn fume particles were 0.17 and 0.10 μm , respectively. Based on this work, the ELPI is identified as a valuable tool for the evaluation of airborne particulate matter in the indoor air of metallurgical production sites. The method is well suited for real-time assessment of morphology (particle shape), particle size, and particle size distribution of aerosols.

Keywords ferroalloy, ferrosilicon, fugitive emissions, particle size distribution, particulate emissions, silicomanganese

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INTRODUCTION

The topic of fugitive emissions in metallurgical plants is important on many levels. First and foremost it is a source of concern regarding the health of the workers.^(1–3) The fumes produced in most metallurgical processes may be harmful if inhaled.^(4–6) Characteristic properties of the particles, especially particle size and chemical composition, influence their impact on human health. The ultrafine and nano-sized particles may, due to their very large surface area be more reactive than the more widely studied, larger particulate matters. They may also behave differently in the respiratory system.^(7–9) As the chemical composition of particles originating from different industries and processes may vary greatly, it is important to characterize the fumes and link them to their origin to enable enhanced risk assessments and enable capture.^(10–12) Moreover, the fumes from industrial plants are a major contributor to the so-called diffuse emissions which are receiving increased attention from authorities as they may influence the air quality of the local, urban communities^(13–15) as well as the environment and climate at large.^(16–18)

A great deal of efforts and investments are currently directed towards improvements in ventilation and filter systems, designed to collect and control the off-gases and fumes from the various metallurgical and mechanical process operations. From the corporate point of view, a process optimized to minimize fume formation would not only improve the indoor air quality and decrease the environmental impact of the plant; it would also minimize material and energy losses associated with the production. To achieve this, more knowledge about the origin and characteristics of the fumes are necessary.

There are several measurement techniques which can be used to characterize and quantify the airborne particulate matter in an industrial plant. The measurements reported on

in this article were performed concurrently with gravimetric filters and laser measurements (LaserDust by Norsk Elektro Optikk AS, Oslo, Norway).⁽¹⁹⁾ The major advantage of the electric low pressure impactor (ELPI, Dekati Ltd., Tampere, Finland) is the detection and quantification of particles as small as 7 nm. The ELPI also enables real-time assessment of the particle size distribution (PSD) of aerosols as well as subsequent analysis of the collected aerosol particles by electron microscopy.^(20–22) The ELPI has previously been used to assess particulate emissions from traffic,⁽²³⁾ combustion,^(24,25) welding fume,^(26,27) and aluminum electrolysis.^(28,29)

This article aims to describe how the novel technology of the electric low pressure impactor can be used to assess indoor air quality surrounding ferroalloy production processes and to establish real-time particle size distributions of aerosols at a ferrosilicon and a silicomanganese production site.

EXPERIMENTAL METHOD

The term “fume,” as used in this article, designates airborne particulate matter (PM) formed by the evaporation of liquid metal. Fume from different locations in silicomanganese (SiMn) and ferrosilicon (FeSi) production plants was collected and analyzed using an ELPI (Dekati Ltd., Tampere, Finland) which is described below. For proper function of the ELPI a particle density should be assumed for the mathematical conversion of the as-obtained electrical current values to a number concentration (number of particles per volume of air). In these studies, the assumed density values were that of amorphous silica (SiO₂, 2.2 g/cm³); this assumption is motivated and discussed in the following sections of this article. The density value has a relatively weak influence on the number concentration as it merely affects the size range intervals for each impactor stage (the *cut-off* values between the different size fractions).

The same assumed density was also used in a second step when the number concentrations were algebraically converted to mass concentrations. The number-to-mass conversion was performed using the filter functions in the ELPIvi software (Dekati Ltd., Tampere, Finland)⁽³⁰⁾ and the density has a large influence on this conversion, which in turn introduces significant error sources as discussed in further sections.

The particles are collected in the ELPI on aluminium foil substrates, one for each size fraction as detailed in Table I. These particle samples were subsequently analyzed by scanning electron microscopy (SEM), transmission electron microscopy (TEM), and energy dispersive spectroscopy (EDS). The amount of fume particles collected by the ELPI was not, however, sufficient to quantitatively analyze the chemical and/or phase compositions by other methods.

In the ferrosilicon plant, a number of dust samples were collected from the off-gas channel above the furnace (where the gas and dust from the furnace top ventilation system are collected and removed) simultaneously to the ELPI measurements. These samples were more substantial and could be analyzed by both EDS and inductively coupled plasma

TABLE I. Aerodynamic Diameter Intervals and Geometric Mean Aerodynamic Diameters

Impactor Stage No.	Aerodyn. diam. range (μm)	Geom. mean aerodyn. diam. (μm)
1	0.007–0.028	0.02
2	0.028–0.054	0.04
3	0.054–0.091	0.07
4	0.091–0.153	0.12
5	0.153–0.259	0.20
6	0.259–0.379	0.31
7	0.379–0.609	0.48
8	0.609–0.942	0.76
9	0.942–1.59	1.22
10	1.59–2.38	1.95
11	2.38–3.97	3.07
12	3.97–9.85	6.25

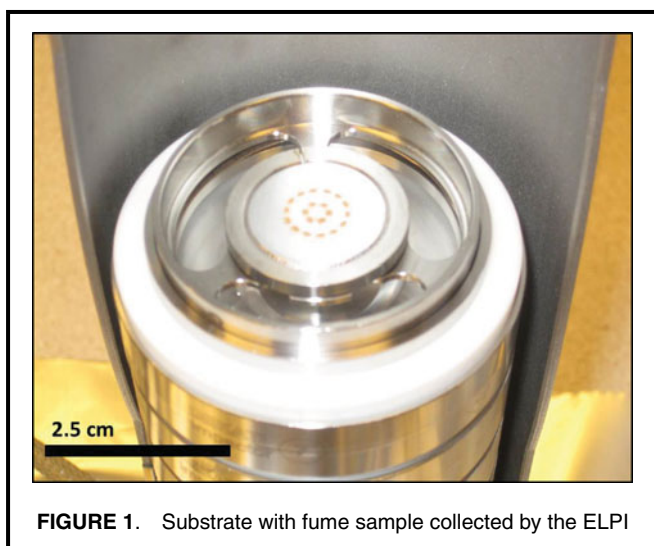
optical emission spectroscopy (ICP-OES). While the samples from the off-gas channel are not directly comparable to the tapping area fume, they may still serve as a reference for the furnace fume of that specific plant.

Dust samples were also collected by gravimetric filters in the roof exhaust of the silicomanganese plant. These samples were used as a reference for diffuse, fugitive, particulate emissions from this specific plant. They were not substantial enough for wet chemical analysis but were analyzed by SEM and EDS in the same way as the ELPI substrates.

The Electrical Low Pressure Impactor (ELPI)

The ELPI classifies aerosols according to their aerodynamic diameter, D_p and collects real-time particle size measurements in the size range of 7 nm – 10 μm; a particle size distribution is established based on the size fractions of the impactor stages. These stages and their aerodynamic diameter intervals with corresponding geometric mean diameter values are detailed in Table I. The ELPI was constructed at the Tampere University of Technology (Tampere, Finland) for the purpose of monitoring particle size distributions of airborne particulate matter. Detailed descriptions of the ELPI function and its principles of operation are given in the literature.^(20–22,31,32) The aerosols analyzed in this study were collected on greased aluminium foil substrates, as shown in Figure 1. This enables subsequent analysis by electron microscopy and energy dispersive spectroscopy but the amount of material collected is too small to allow more quantitative and/or phase compositional analysis.

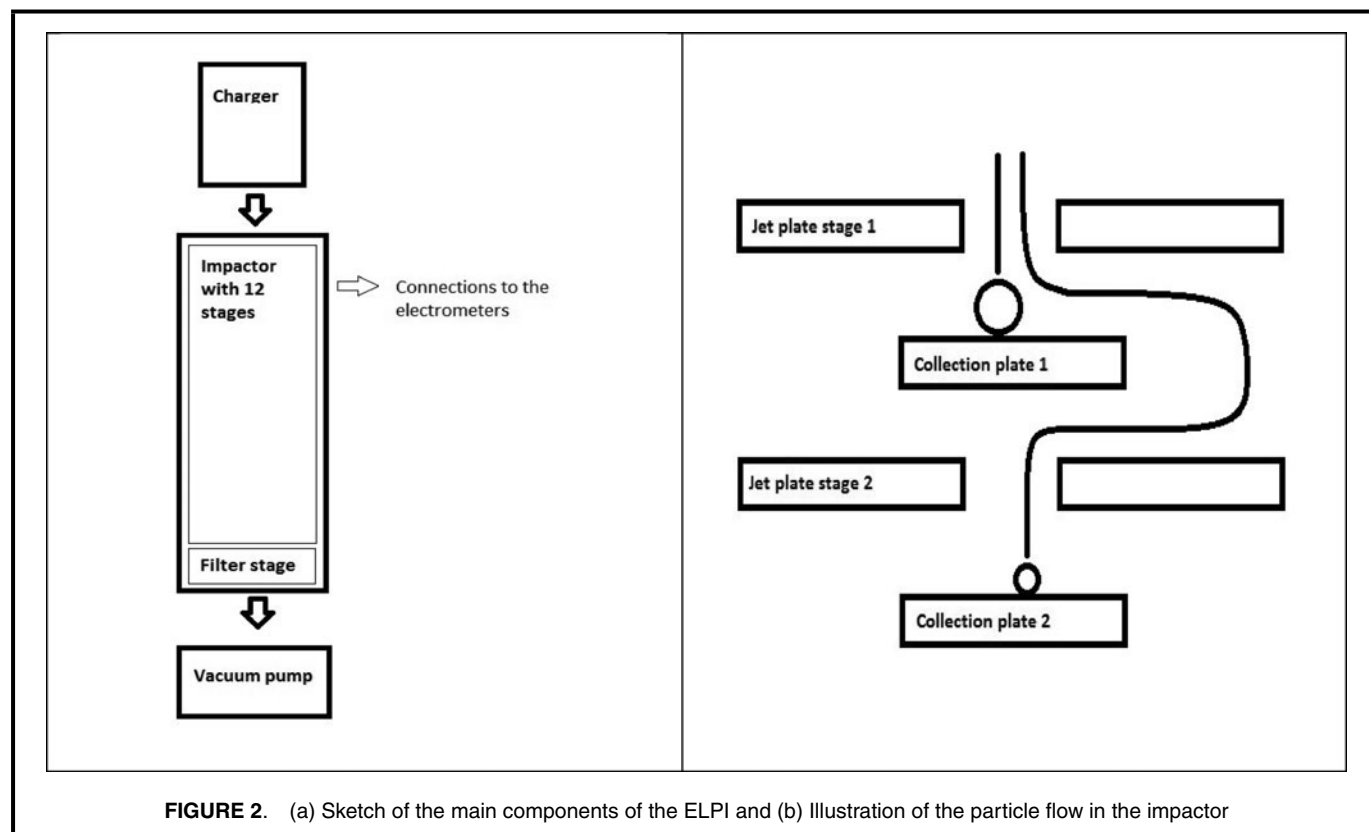
The operating principle of the ELPI is illustrated in Figure 2a; it includes particle charging in a corona charger; inertial classification in a cascade impactor; and electrical detection of the collected particles by a multichannel electrometer. A vacuum pump is used to control the airflow (10 l/min) through the instrument.

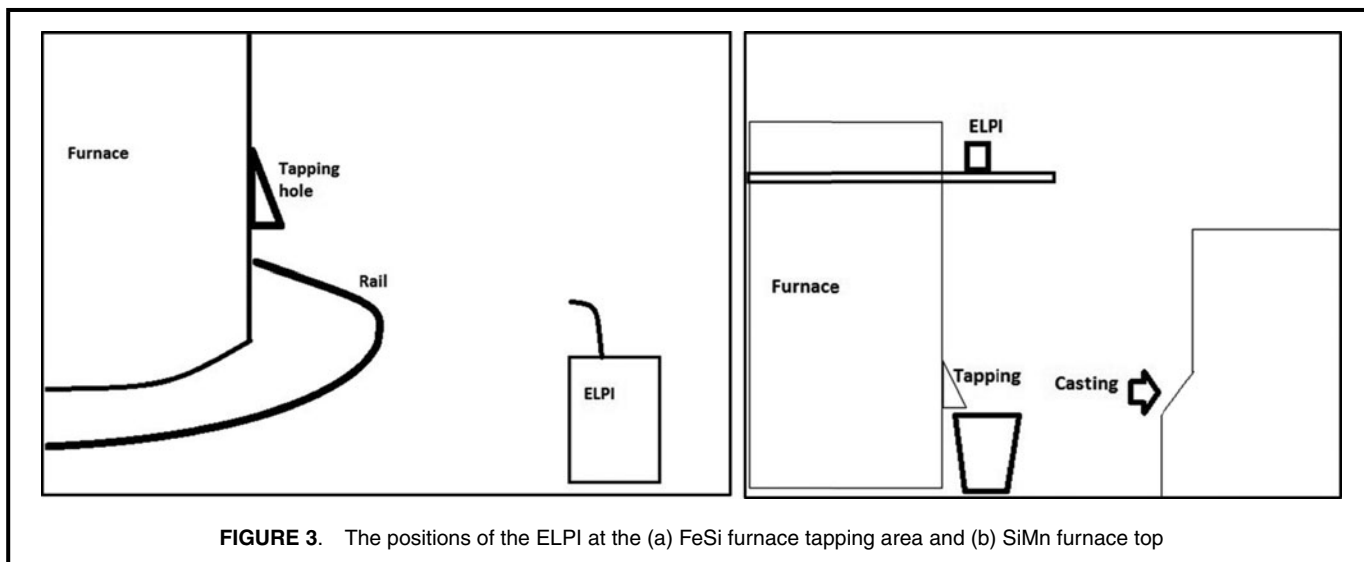


The particles enter the ELPI by use of the vacuum pump. They pass through the charger unit where the particles become electrically charged. The particles then enter the impactor unit which consists of 12 stages and a filter stage. Each impactor stage is connected to an electrometer. The particles are then separated into different stages depending on their aerodynamic behavior. As a charged particle is collected in an impactor stage, it produces an electrical current recorded by the electrometer.

The impactor classifies the particles according to their flow inertia. Figure 2b illustrates the collection of particles from the air stream in the impactor. The air stream passes through a nozzle in a jet plate upon entering each impactor stage. After the nozzle, the stream makes a sharp turn which allows the air to pass between the jet plate and the collection plate. The streamlines are bent just above the collection plate. The large particles, which cannot turn so abruptly, will be deposited on top of the collection plate. Smaller particles are more agile and will therefore follow the air stream to the next stage. The radius of the streamlines decreases for each impactor stage. Each stage has a certain probability for errors, such as losses of particles due to diffusion and bounce-off effects. The instrument signal is mathematically treated to correct for these errors.^(31,33)

In an impactor, the size fractionation of aerosols is based on the aerodynamic diameter (D_p). It is defined as the diameter of a unit density spherical particle having the same settling velocity as the actual particle. Another widely used definition is the Stokes diameter (D_s), which is the diameter of a spherical particle having the same bulk density and settling velocity as the actual particle.⁽²⁰⁾ The aerodynamic diameter is the output parameter of the ELPI measurements, and the Stokes diameter is used to calculate the cut-off values between the different size fractions. Naturally, most aerosols are not perfectly spherical and the morphology (particle shape) of the particles and/or particulate agglomerates may vary greatly, which in turn will





influence their aerodynamic properties through their inherent inertia.

The Ferroalloy Plants

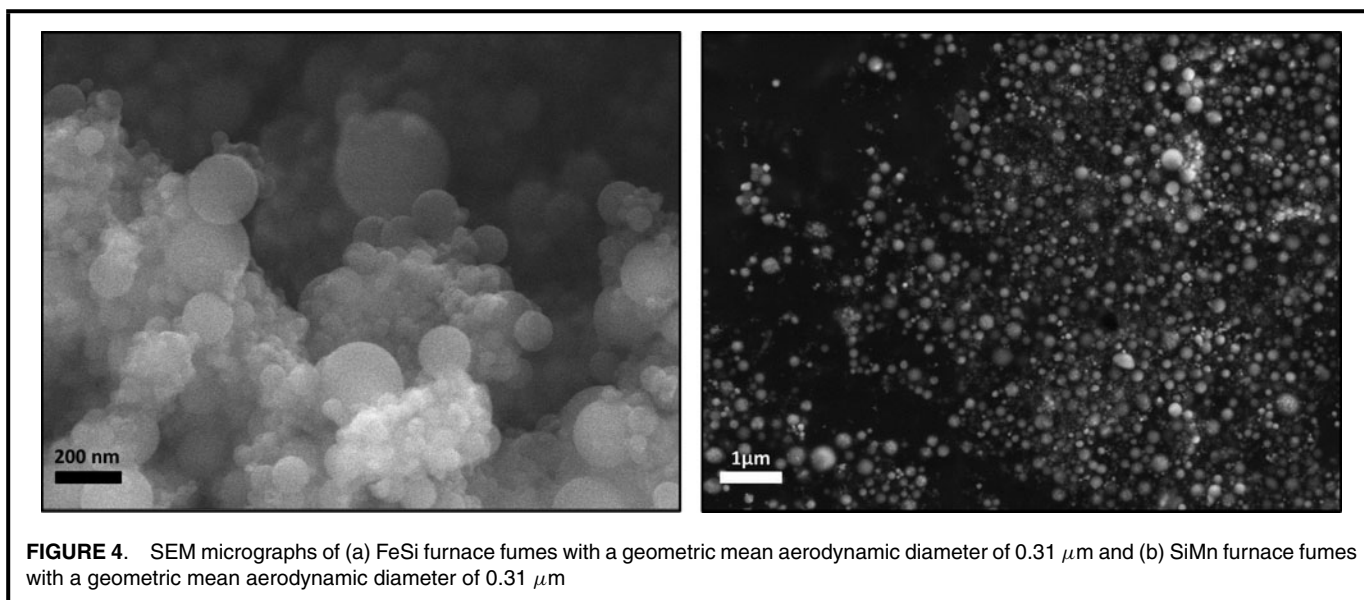
The alloys ferrosilicon and silicomanganese are both produced in electric arc furnaces. Raw materials include crushed or pre-sintered minerals and ores, and sometimes scrap iron or slag from ferromanganese production, in addition to carbonaceous reductants (coke and coal). The raw materials are fed at the top of the furnace. Inside the furnace, the raw materials react and form a raw metal which is tapped from the furnace into a transportable ladle. The tapped metal is generally refined in the ladle by a slag process.⁽³⁴⁾ The refined metal is then cast and crushed to the customers' specification. A thorough description of the production processes for FeSi

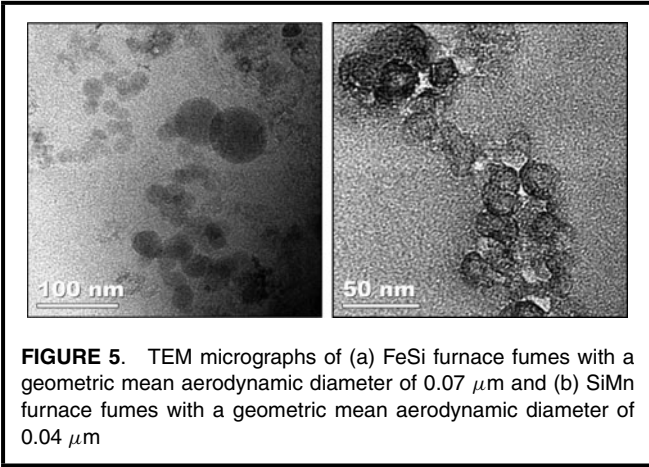
and SiMn can be found in the textbooks by Schei⁽³⁵⁾ and Olsen,⁽³⁶⁾ respectively.

The major source of fume is the furnace itself, yet modern furnace design in combination with powerful ventilation and filter systems are efficiently removing most of the furnace top gases and fumes. Typically, the tapping, refining, and casting operations are the main sources of fume inside the plant and of the so-called diffuse emissions from the plant.⁽³⁷⁾

In the FeSi plant, the ELPI was set up in the furnace tapping area (see Figure 3a) and the duration of the measurements was approximately 3 hr. The FeSi furnace was tapped continuously and substantial gassing and fuming from the tapping hole was observed during the measurement.⁽³⁸⁾

In the SiMn plant, the ELPI was set up on the top furnace floor (see Figure 3b). The ELPI measurements were performed inside and outside a fume collecting curtain to assess the effect



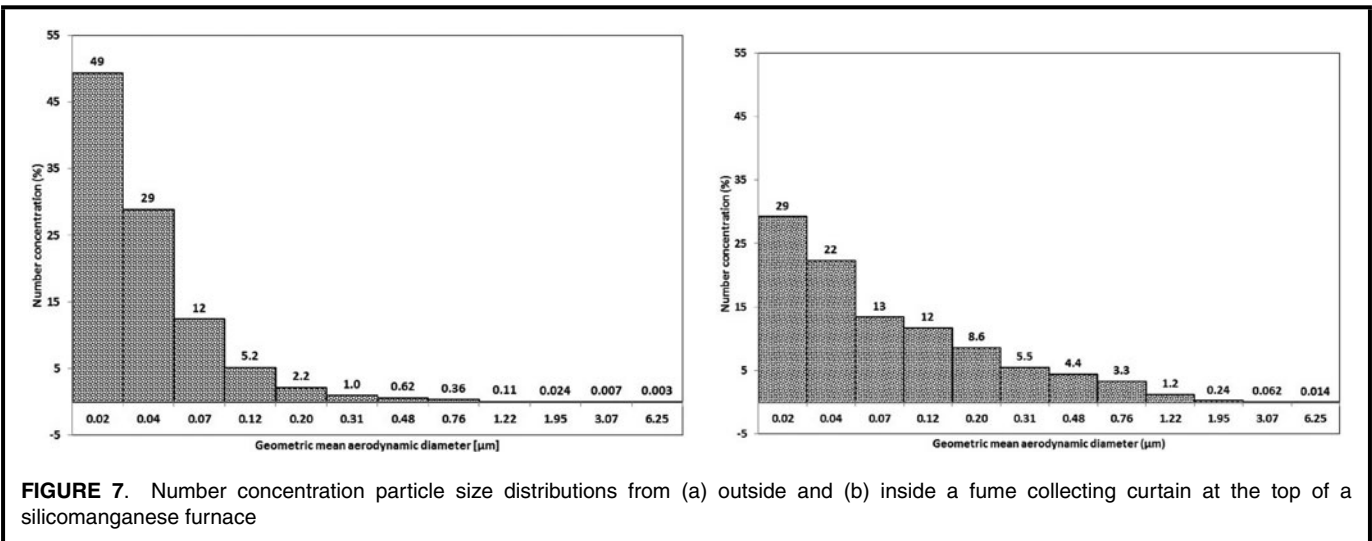
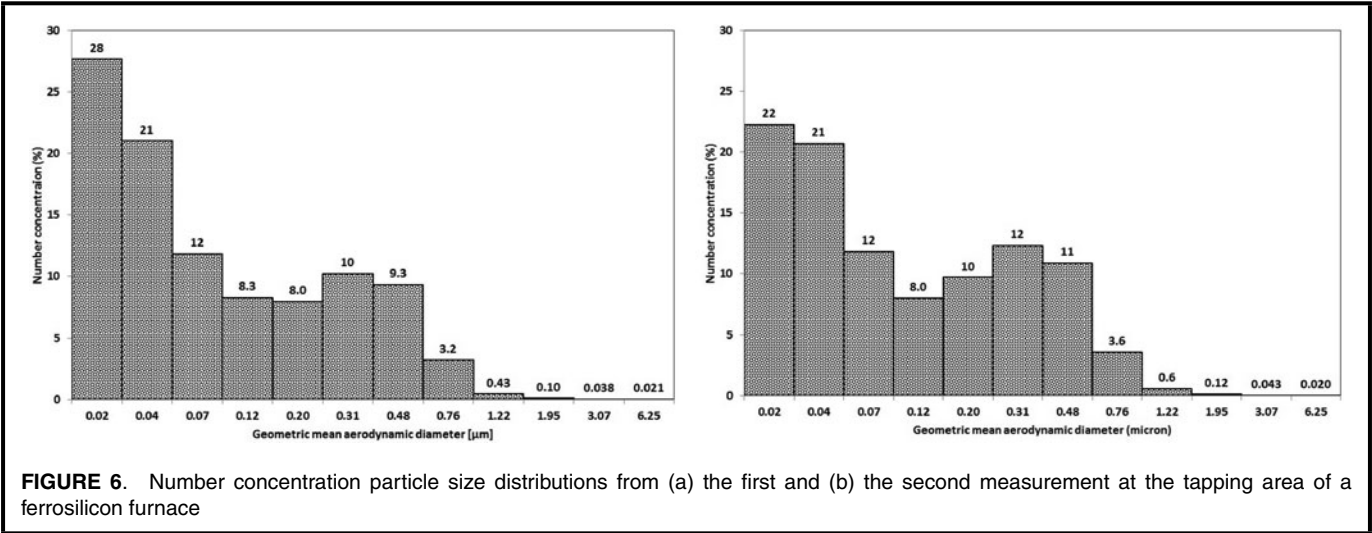


of the curtain on the indoor air quality as experienced by the workers. The duration of the measurements shown was approximately 3 hr.⁽³⁸⁾

RESULTS AND DISCUSSION

Scanning electron micrographs of the FeSi and SiMn fumes are shown in Figure 4. The particles are from the ELPI stages which have geometric mean aerodynamic diameters of $0.31 \mu\text{m}$ ($0.259\text{--}0.379 \mu\text{m}$). The morphologies of the particles in both plants were corroborated by SEM analysis of off-gas fume and roof exhaust fume, respectively.

The particles consist of agglomerated spheres and this morphology is typical for particles in the size range from 0.09 to $0.94 \mu\text{m}$. The presence of agglomerates in FeSi fume has previously been reported by Dingsøyr et al.⁽³⁹⁾ They applied the notions of “primary” and “secondary” agglomerates to



describe the fume particles; the former being particles where the protoparticles (in our case, the spheres) are held together by material bridges and the latter being primary agglomerates attached to each other by weak van der Waals forces. Secondary agglomerates are not likely to be recorded by the ELPI as they may disengage in the charger unit. Primary agglomerates on the other hand will remain agglomerated and it is therefore the primary agglomerate sizes, and not the spherical protoparticles sizes, which are recorded by the ELPI. Hence, the average particle diameter recorded by the ELPI differs from particle size measurements by SEM and Brunauer-Emmett-Teller (BET) surface adsorption,⁽⁴⁰⁾ whereas it corresponds well to the values obtained by accelerated sedimentation.⁽³⁹⁾

Transmission electron micrographs are shown in Figure 5. The micrographs show particles from the ELPI stages with geometric mean aerodynamic diameters of 0.07 and 0.04 μm , for the FeSi and SiMn fumes, respectively. These results are representative for the particles collected in the stages with size ranging from 0.03 to 0.09 μm . The TEM analysis shows that the spheres in Figures 5a and 5b are amorphous. No TEM results were obtained for the particles in the smallest fraction due to the difficulties associated with TEM sample preparation.

The EDS elemental analysis, summarized in Table II, specifies that the fume from the FeSi furnace is dominated by Si and O which indicates that the spheres likely consist primarily of SiO_2 . The fume from the FeSi furnace also includes significant levels of Fe, K, Al, Mg, Na, and Ca, which is in good agreement with the chemical composition of the off-gas fume collected simultaneously at the plant and also with FeSi fumes observed by others.^(39,41) The smallest fractions, analyzed by TEM, had lower levels of trace elements than the larger fractions analyzed in SEM. For FeSi, Fe was the only “contaminant” detected in the fume and Fe may be included in the Cu-grid used as sample holder (C and Cu are the main constituents of the sample holder).

The chemical composition of the SiMn fume particles is more complex and varying than the FeSi fume, which was also corroborated by the gravimetric filter samples collected

TABLE II. Elemental Analysis of the Fumes as Obtained by EDS

	FeSi fume	SiMn fume
Major elements	Si, O	Si, Mn, O
Minor elements	Fe, K, Al, Mg, Na, Ca	Mg, Ca, Al, K
Trace elements		Na, Fe, Zn, Cu, Cl

simultaneously at the plant. It consists primarily of Si, Mn, and O. Certain size fractions, most notably those with aerodynamic diameters larger than 1 μm , contain significant levels of Mg, Ca, Al, and K and also traces of other contaminants such as Na, Fe, Zn, Cu, and Cl. The relatively high levels of alkali (Na and K) are also found in laboratory scale experiments which will be published elsewhere. The SiMn fume particles morphology is very similar to the FeSi fume. The primary units of the particles are spheres which tend to agglomerate. The morphology and chemical composition of the fume seem to be in fair accordance with previous reports.^(42, 43)

For SiMn fume, the smallest fractions display greater dominance of Si than the larger fractions and the ultrafine particles contain significantly lesser amounts of Mn and trace elements than the larger-size fractions. Such findings have also been reported for Mn-containing welding fumes.⁽²⁶⁾ It is also possible that the ultrafine particles originate from sources outside the fume curtain; as this particular SiMn plant is hosting other ferroalloy processes, which are only partially separated from the SiMn furnace hall, it is possible that some intermixing of particles through air exchange has occurred.

Figures 6 and 7 show the particle size distributions (PSD), in terms of number concentration, as a function of the aerodynamic particle diameter, D_p , as obtained by the ELPI. The number concentration PSDs are dominated by the smallest-size fractions. The average aerodynamic diameters of the fume particles, as recorded by the ELPI, are 0.17 and

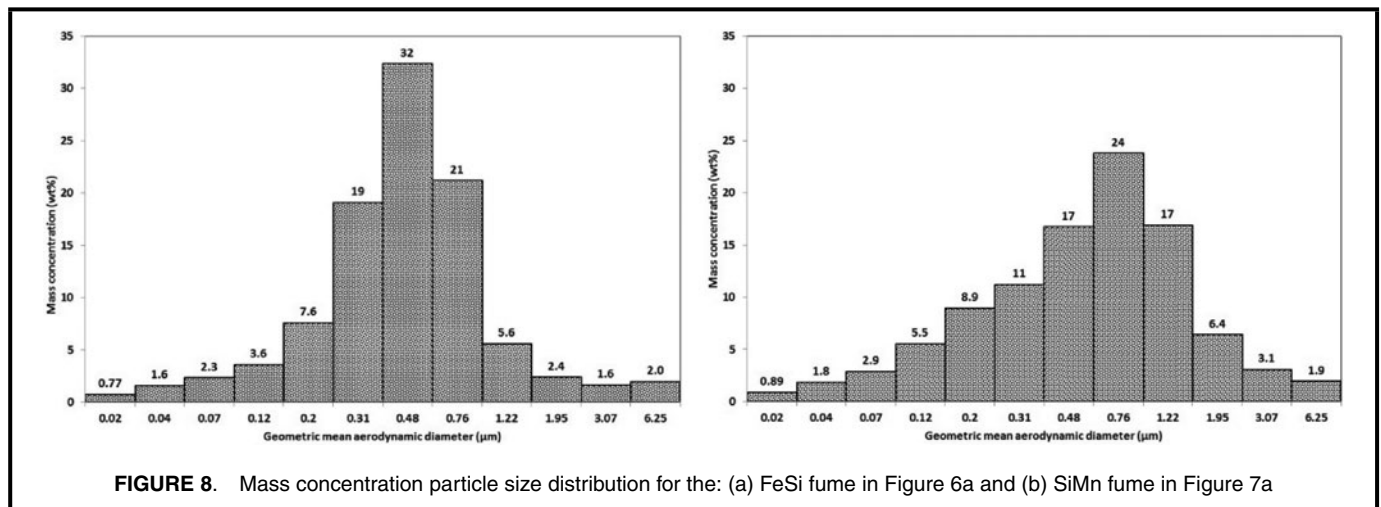


FIGURE 8. Mass concentration particle size distribution for the: (a) FeSi fume in Figure 6a and (b) SiMn fume in Figure 7a

0.10 μm for the FeSi and the SiMn fume particles, respectively. Using the terminology applied by Preining,⁽⁴⁴⁾ the particle diameter can be used to classify aerosols as “fine” ($D_p < 1 \mu\text{m}$), “ultrafine” ($D_p < 100 \text{ nm}$), and “nano-sized” ($D_p < 30 \text{ nm}$). For the FeSi particles 99% (percent by numbers) are so-called fine aerosols; 58% are ultrafine aerosols and 25% are nano-size aerosols. For SiMn fume, almost 100% of the particles collected are fine aerosols; 78% are ultrafine and 40% are nano-sized.

The graphs in Figures 6 and 7 show that the two PSDs taken at each plant are relatively similar in shape. Still, two measurements at each site are not statistically sufficient to assess the characteristics of the airborne particulate matter in the plant. To assess the temporal variation of the indoor air quality and potential diffuse emissions of a plant multiple and frequent measurements would have to be undertaken. The PM should also be evaluated at different sites within the plant. This work shows that such a project is feasible, by use of an ELPI, although it is beyond the scope of this particular study.

In Figure 8, the same particle size distributions as above are recalculated as mass concentrations. From the EDS results, the fume from both furnaces was found to be mainly SiO_2 and therefore a density of 2.2 g/cm^3 was assumed in the mass concentration calculations. For the SiMn, however, this is not an ideal assumption as the chemical composition of the particles from the SiMn production is quite diverse and the phase composition remains largely unknown.^(42,43)

It is, however, a useful approximation to compare the number concentration distributions (Figures 6a and 7a) with the mass concentration distributions (Figures 8a and 8b), since most administrative standards for airborne particulate matter are given in mass concentration units. For the FeSi furnace, the percentages by mass are as follows: 88 wt% are fine, 5 wt% are ultrafine, and merely 1 wt% are nano-sized. For the SiMn furnace these numbers are 72 wt%, 66 wt%, and 11 wt%, respectively.

CONCLUSION

The electric low pressure impactor has been successfully applied to assess the particle size distribution of fume particles in the indoor air surrounding two ferroalloy production processes. The aerosol sizes recorded by the ELPI correspond to the aerodynamic diameters of the so-called primary agglomerates. Particle size distributions in the range of 7 nm to 10 μm have been established for both sites. The majority of the particles are smaller than 1 μm and approximately 5 wt% are so called ultrafine aerosols.

The samples collected by ELPI were not substantial enough for in-depth chemical analysis. Elemental analysis was therefore limited to EDS. The morphology of the collected particles was evaluated by SEM and TEM. The fume consisted primarily of spherical, amorphous silica protoparticles with trace levels of alkali and other contaminants.

This work shows that the ELPI has great potential use for the assessment of airborne particulate matter from ferroalloy

production. For a thorough PM assessment, multiple and frequent measurements should be carried out at different sites of the plant.

Number concentration particle size distributions were measured in-situ by the ELPI and mass concentration particle size distributions were calculated from these results. The differences between the particle size distributions based on number and mass concentrations are significant and may be attributed to the very small masses of the ultrafine and nano-sized particles.

SUPPLEMENTAL MATERIAL

Supplemental data for this article can be accessed at tandfonline.com/uoeh. AIHA and ACGIH members may also access supplementary material at <http://oeh.tandfonline.com/>.

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