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Research Paper

Artisanal ceramic factories using wood combustion: A nanoparticles and human health study

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ABSTRACT

The ceramics industry, resulting from developments of modern compounds, is a segment of great influence in worldwide sustainability. Artisanal ceramic factories based on wood combustion have significant risks for the creation and discharge of atmosphere nanoparticles (NPs) and ultra-fine particles (UFPs). At present, there is insufficient recognition on the influence of engineered-NPs on the atmosphere and health. Real improvements are indispensable to diminish contact with NPs. The present study demonstrates the main NPs and UFPS present in an area of intense artisanal wood-combustion ceramic manufacturing. Particulate matter was sampled for morphological, chemical, and geochemical studies by sophisticated electron microbeam microscopy, X-Ray Diffraction, and Raman spectroscopy. From NPs configuration (<10 nm) we identify nucleation. Several amorphous NPs (>10 nm) were produced around the studied artisanal ceramic factories. This study presents an indication of the recent information on population and work-related contact to NPs in the artisanal ceramic factories and their influence on health.
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1. Introduction

For millennia, the ceramic manufacturing has produced an extensive variety of products with influence on human culture and sustainability. Mostly, a ceramic compound can be described as an inorganic, heat-resistant material constituted of both metallic and non-metallic compounds. Ceramics have a vast utilization and are applied in numerous engineering manners and modern human advances. Bricks, ceramic tiles, drainage pipes, tableware, and decorative ware are several of their most modern functions (Bessa et al., 2020). They are utilized to fabricate an array of provisions such as cutting instruments, coats, microelectronic apparatus, and medicinal commodities (Medina et al., 2011). An important quantity of the world's ceramic productions is situated in the America's, with many employees and a projected fabrication value of billions of dollars every year. In the ceramic industrial activity, the compounds utilized can go through many processes that can produce diverse dangers from the health and ecological systems (Salmatonidis et al., 2018). For example, atmosphere discharges in the ceramic manufacturing characterize an ecological concern due to the liberation of NPs and UFPs or powder in management and managing of raw constituents (Bessa et al., 2020). Subsequently, many engineering practices which cause NPs discharges have been of mounting consideration in last two decades (Dotto et al., 2011, 2012, 2015a, 2015b, 2016; Peres et al., 2018; Rodrigues et al., 2018).

Most of the ceramics produced in Santa Catarina state, Brazil, use wood combustion without proper care for health and/or the environment. Wood burning produces a significant amount of ambient particulate matter and is an additional concern regarding the workers and population (Vicente et al., 2020). But, the influence of this discharge on atmosphere contamination may be at a complex level. Numerous works on air contamination from cooking with wood fuels describe NPs and UFPs intensities generally surpassing the World Health Organization (WHO) atmosphere property standards (Bartington et al., 2017). Several references have described the character of atmosphere contamination ascending from wood combustion on numerous respiratory (Raspanti et al., 2016; Vicente et al., 2020) and non-respiratory disorders, such as child death, cardiac infection, nasopharyngeal cancer, and melanoma (Wylie et al., 2014).

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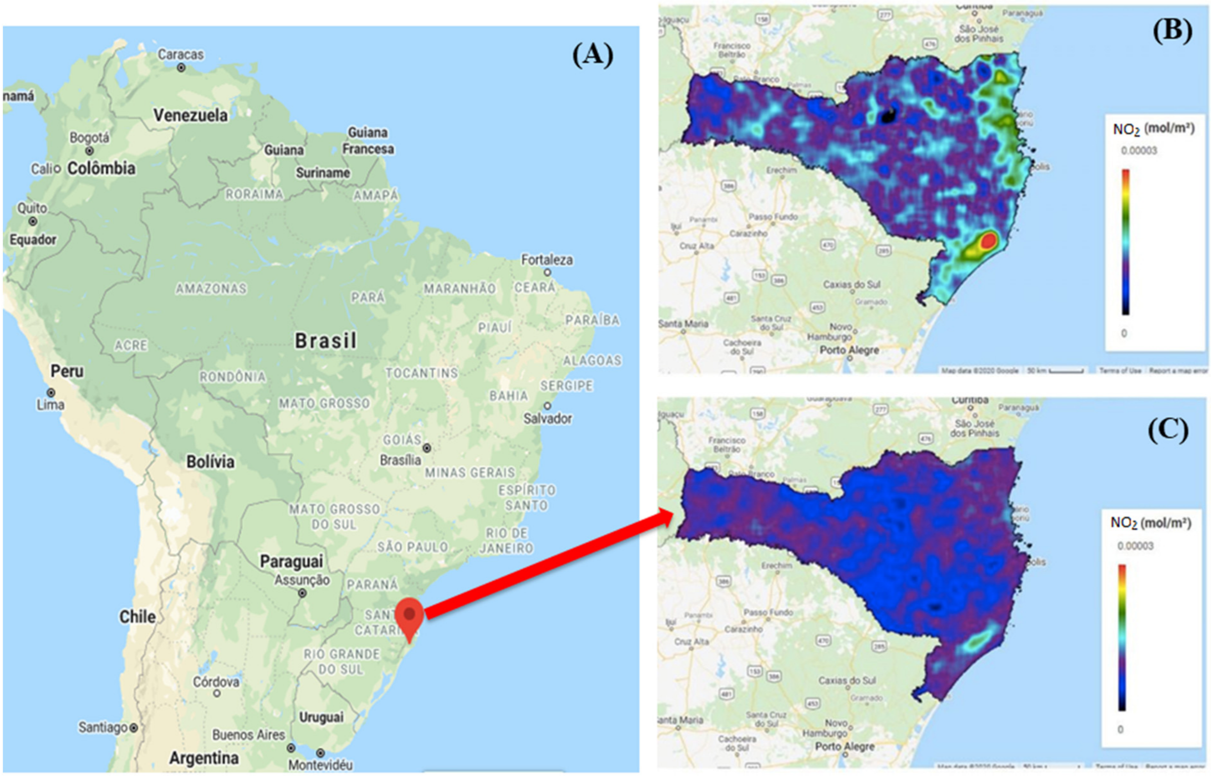

Fig. 1. (A) Studied area; (B) NO_2 distribution in Santa Catarina state before the COVID-19 pandemic; (C) NO_2 distribution in Santa Catarina State in March 2020.

Santa Catarina (Fig. 1) is one of the most developed regions in southern Brazil. The appropriate environmental political bodies poorly supervise industries that work with coal, ceramics and civil construction, agriculture, clothing, among others. Of these activities, only the bibliographic references about the coal-fired power plants are listed (Silva et al., 2009; Quispe et al., 2012; Oliveira et al., 2013; Cutruneo et al., 2014; Dias et al., 2014; Rodriguez-Iruretagoiena et al., 2015; León-Mejía et al., 2018; Nordin et al., 2018; Duarte et al., 2019; Gredilla et al., 2019).

In this study, this is considered as an opportunity to present the environmental assessments regarding atmospheric contamination, as well as providing references to the appropriate environmental authorities of the area under study. The study and description of UFPs and NPs have been related with respiratory health results through successions of important life devices (Agudelo-Castañeda et al., 2016, 2017; Stone et al., 2017; Oliveira et al., 2020; Silva et al., 2020a).

This study aims to identify the NPs under real-world-operation circumstances in an artisanal ceramic production area. The methodology describes the chemical and morphology of NPs. Thus, the present study aims to increase the existing scientific information on contact mitigation policies by giving qualitative evaluations of efficacy of precise procedural actions.

2. Sampling and study area

Germans, Poles, Portuguese, and Italians, who boosted the development of this region, massively colonized the south of the state of Santa Catarina over 100 years ago. These colonizers brought some practices that are potentially hazardous from the environmental point of view and for human health, such as the use of energy through a coal-fired power plant, production of ceramics without due care for health, family farming with extensive use of agrochemicals, steelmaking, among many other activities. Fig. 2 illustrates representations of artisanal ceramics that have been operating for more than half a century. In some cases, it has always used artisanal labor, but, in a few cases there is also the presence of machines such as tractors. In no case was there any use of personal protection equipment and excess wood combustion was always used.

Several studies, in particular, indicate the activities related to the extraction, transport, and use of coal as being the main source of contamination of this region (Ramos et al., 2015; Civeira et al., 2016a, 2016b; Dalmora et al., 2016; Oliveira et al., 2019a). Studies on ceramic microfactories in cities such as Treze de Maio, Morro da Fumaça, and others have not been reported. This study will help the state's environmental agencies and will also serve as a reference for other countries that have artisanal ceramics using wood combustion, such as China and India.

Such factories use wood combustion and occasionally generate significant fires in the region (Fig. 3A–D), and a high risk to the health of their workers and population. In private meetings with politicians and environmental agencies in the state of Santa Catarina, it was possible to confirm the great deficiency of air quality monitoring stations for the area under study and for the entire state. The low scientific production of local universities regarding the implications of air contamination on the health of the population is also a strong aggravating factor.

Through surveys conducted in 2017, with questions focused on the ceramics of the region under study, it was possible to claim that the population is aware that it is exposed to the contaminants emitted by such activity, especially due to allergies, coughs, and increased respiratory problems in the winter, as smoke from chimneys (see Fig. 3A) tend not to disperse as easily as at other times of the year. Despite this, no previously reported studies were found on the manufacture of ceramics using wood combustion. In view of this scenario, the owners of the houses were asked to authorize the installation of a low-cost, self-made passive sampler (LSPS) consisting of a PVC tube with one interior

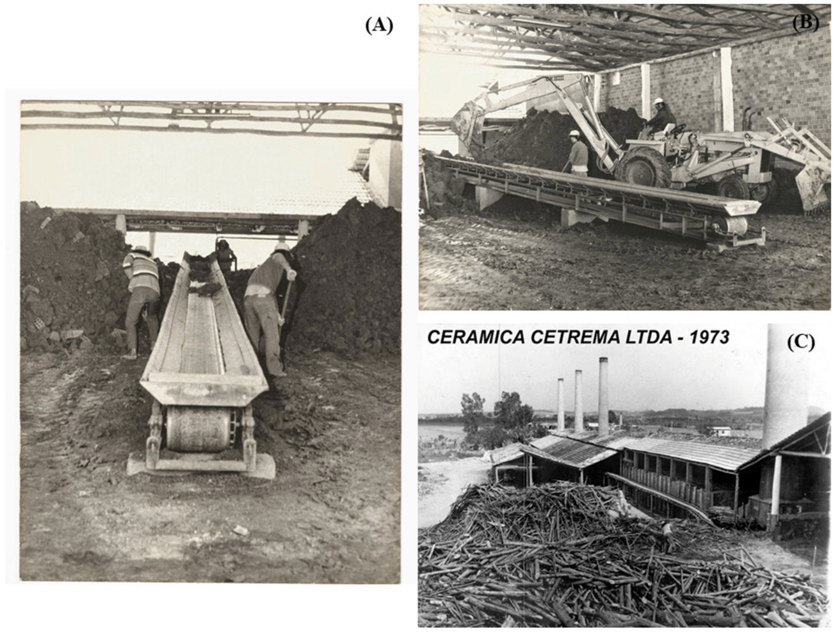

Fig. 2. Illustrative photos of the artisanal ceramic factories in this study. (A) Historical illustration of artisanal labor; (B) use of machines without individual protection equipment; (C) use of excess wood combustion.

pin stub covered with C-tape (Cortés et al., 2020; Liu et al., 2020; Silva et al., 2020b) in each affected house, especially considering the predominant wind direction of the last five years. Forty-six samples were sampled in 2018 and 2019 to evaluate both the PHEs in proportion to the NPs and their overall levels of occurrence within the ceramic production zone. The LSPS, previously described by Silva et al. (2020a), were mounted in the sampling areas (Fig. 1A) and were utilized to directly accumulate PM of multiple sizes and shapes.

Fig. 3. (A) Actual ceramic production; (B, C and D) fires problems.

In March 2020, it was reported by state authorities that NO_2 contamination across the state of Santa Catarina has been reduced to less than half of what it usually is in that same period in other years. In Fig. 1C, it is not even obvious that this atmospheric contamination stopped with the COVID-19 pandemic; the coal-fired power plant continued to operate and the artisanal factories are family operations without crowds of people, so almost all continued in full operation. Fig. 1B and C helps us to see that the region under study is the most critical in the state for atmospheric contamination, both in normal years and in 2020 due to the pandemic. The ongoing COVID-19 pandemic has already spawned studies of the synergy between the virus, underlying/pre-existing health issues, and intrinsic (such as smoking) and extrinsic (such as industrial pollution) air-quality issues (Conticini et al., 2020; Dantas et al., 2020; Fattorini and Regoli, 2020) and will certainly generate decades worth of expanded studies.

3. Materials and methods

To optimize time and resources, 150 particles were studied in each sampled house. Of these, field-emission scanning electron microscopy (FE-SEM) was used to study fifteen 10–20-μm particles, thirty 5–10-μm particles, thirty 1–5-μm particles, and thirty 100–1000-nm particles and high-resolution transmission electron microscopy (HR-TEM) (Silva et al., 2009; Hower et al., 2013; Ferrari et al., 2019; Dalmora et al., 2020; Gómez-Plata et al., 2020) was used to study 45 particles smaller than 100 nm. For this study, a Cu-grid of HR-TEM in the LSPS was also added for the analysis of NPs by HR-TEM. Such a procedure does not require sample preparation prior to analysis by advanced microscopy (AM), Raman Spectroscopy (RS), or X-ray powder diffraction (XRD). This results in more realistic results since they do not alter the nature of the sampled particles.

The solid fraction was analyzed with a Bruker X-ray diffractometer (model D8 DISCOVER) with NAP-LOCK X-ray optics to examine mineral and amorphous phase occurrences (Ramos et al., 2014, 2017, 2019; de Vallejuelo et al., 2017; Sánchez-Peña et al., 2018; Gallego-Cartagena et al., 2020; Gómez et al., 2021). Working conditions were as follows: slit fixed at 12 mm, Cu Kα monochromatic radiation, and a 20-mA current at 40 kV. Samples were scanned at a speed of 0.3° 2θ/min (5°–65°). The UNPs samples with mixed compounds were studied by X-ray diffraction (Gasparotto et al., 2018) and a 200-keV high-resolution transmission electron microscope (HR-TEM) equipped with a FE cathode and an energy omega filter for high accuracy of the assembly and atomic arrangement, fast Fourier transform (FFT), microbeam diffraction (MBD), selected area electron diffraction (SAED), scanning transmission electron microscopy (STEM), and energy-dispersive X-ray spectroscopy (EDS) (Oxford Instruments INCA 4.09 software). The chemical components with high atomic numbers appear in the brightest areas of the identified image and those with low atomic numbers are present in the dark-field areas. The AM/EDS reveal that the PM has slightly diverse geochemical conformation (Rodriguez-Iruretagoiena et al., 2015). Before microscopic analysis, the specimen holder was cleaned with an advanced plasma system (Gatan Model 950) to minimize contamination. Even so, a blank was analyzed at the same time as the samples in order to ensure that the samples do not contain impurities from the specimen holder. The FE-SEM model Sigma 300 VP (Carl Zeiss, England) with field emission filament (FEG- Field Emission Gun) of the Schotky type (tungsten filament covered with zirconium oxide) and equipped with a Gemini column (Zeiss, England). The images were obtained using the secondary detector (SE2) in high-vacuum mode (1×10^{-9} bar); working distance of 5 mm; opening of 20 μm; and magnifications of 1000×, 2500×, and 5000×. The energy-dispersive X-ray spectrometer (EDS) spectra were generated by the EDS X-ray detector (model Quantax 200-Z10, Bruker, Germany) equipped with a 10-mm^2 quartz window and ESPRIT software. The EDS were obtained using the secondary detector (SE2) in the variable-pressure mode, which consists of the insertion of N_2 gas in the sample compartment, allowing the variation of the partial pressure between 1 Pa and 133 Pa (1 Pa = 1×10^{-5} bar), working distance 8.5 mm, energy of 20 kV, and opening of 60 μm (Ribeiro et al., 2010, 2013a, 2013b; Cerqueira et al., 2011, 2012; Dias et al., 2014; Zamberlan et al., 2020). HR-TEM equipped with EDS was utilized. The X-Ray diffraction (XRD) pattern after amorphous extraction of obtained samples were acquired with Cu-Kα radiation (λ = 1.54 Å). Raman Spectroscopy was utilized to study the particles molecular organization (Silva et al., 2009).

4. Results and discussion

In the studied area, ceramic companies are important for the livelihood of families in the region. On the other hand, not only the workers of the ceramic companies but all the local population has their health impaired due to the lack of concern of the elected officials and to legislation that is not rigid and active regarding the small-scale ceramic factories that use wood combustion. Such facts have resulted in a chaotic social problem and little control. Given the results of NPs and UFPs obtained in this study, it is not surprising that even in the COVID-19-pandemic season, a time of diminished pollution in many corners of the world (Fattorini and Regoli, 2020), atmospheric contamination has not disappeared in southern Santa Catarina (see Fig. 1B and C). Even though the coal-fired power plant has been investing in solar energy, such an investment does not yet represent 2% of the total energy generated by the company. As the ceramic factories are relatively far away from urban centers and because they are small-scale productions, they have not stopped operating during the COVID-19 pandemic, so it is evident that the NO_2 results are especially due to these two anthropogenic activities. There is legitimate concern that when the pandemic ends, such industries can use this fact to increase their activities to “recover time” lost during the economic slowdown, resulting in further contamination of the area under study, consequently affecting the health of workers and the population.

Bessa et al. (2020) confirmed the possibility of inhaling NPs and UFPs in modern, well-regulated ceramic factories. In artisanal factories and in the area under study, the importance of reporting the NPs is quite evident. The data present in this study will serve as a reference for other countries such as China, India, among others where there are handcrafted ceramics. Of the 150 particles detected in each sample, on average, less than 11 appear to come from the coal-fired power plant, while between 50 and 70 were totally isolated, and it can be proved that they came from wood-fired-combustion or from the transformation of clays by heating (alterations such as dehydration of clays, nucleation, sulfates and oxides formation, among many others). The remaining particles (that is, almost 50%) were complex mixtures of all the sources in the area. For a better understanding, the three main sources of NPs and ultra-fine particles detected in this study are described below.

As illustrated in the graphical abstract, some ceramic factories are located between rice fields. The evident contamination by UFPs and NPs is a worrisome reality in the study area. Aware that many of the NPs represent rice contamination, the rice trade is highly affected especially due to the organic contaminants generated by wood combustion. In view of this scenario, it is important that future studies are carried out to assess the level of contaminants in rice growing.

The abovementioned works support the significance of examining UFPs and NPs process-produced discharges in studied activity workplaces and their influence on employee atmosphere contact. Overall, the described reflections and results highlight the significance of the hazard evaluation and the application of anticipation methods to increase business atmosphere characteristic in studies ceramic area.

4.1. Coal and coal by-products particulate matter

Although it is not the main objective of the present study, it is necessary to report the presence of NPs and UFPs derived from the transport, use, and combustion of coal in the area under study. This is because, despite the approximate distance of 25–30-km from the coal-fired power plant, as the predominant wind direction varies according to the season and also from year to year (considerable variations were noted between 2018 and 2019) particles, including pyrite (Fig. 4), jarosite, sphalerite, and marcasite derived from the coal, can reach long distances, including that of the study area. Neither the soil in the region under study nor the material used in ceramics contain such compounds. Previous studies on coal from Santa Catarina (Gasparotto et al., 2018; Duarte et al., 2019) have reported the existence of such minerals.

According to information from the coal-fired power plant, electrostatic precipitators can capture up to 99% of particles up to 10 µm. No data were found on the installation of electrostatic precipitators when the coal-fired power plant was built. The impact on the region can be historic, especially for particles larger than 10 µm. According to several studies on this coal-fired power plant, the soil of the region contains particles of coal fly ash smaller than 10 µm (Oliveira et al., 2014; Rodriguez-Iruretagoiena et al., 2015; León-Mejía et al., 2018; Gredilla et al., 2019). This fact suggests that some of the detected micro and nano-spheres (Fig. 5) probably come from the coal-fired power plant. This creates an additional risk for the population of the area under study, after all, such spheres, according to the analysis of EDS, contain elements such as As, Hg, Cd, and Pb, among others that are retained in the fly ashes during the coal combustion (Sehn et al., 2016; Dutta et al., 2017, 2020; Oliveira et al., 2018a; Oliveira et al., 2018b; Saikia et al., 2018; Silva et al., 2020c). Ninety-six percent of the ultrafine spheres that are likely to be coal-derived fly ashes are composed of major elements present in coal (EDS from Fig. 5). In general, detected ultra-fine spheres (<1 µm) contained a higher proportion of metals than the other groups (discussed in the next two sections). This may be because the temperature in the coal-fired power plant is higher than in wood combustion, and to using an extremely oxidizing environment to obtain the maximum energy from the coal utilized. As a result, organic matter is more likely to be combusted in a coal-fired power plant than the wood burned in ceramic factories. Plus, there is a fundamental difference between wood and the bituminous coal burned in the power plant.

4.2. Ceramic and wood particles

At present, few studies on NPs contact in the ceramic activity have been published. Most of ceramic raw compounds are in powder form. Consequently, when handling these compounds, principally in organization, transportation, accommodation, and management processes, UFPs resuspensions are introduced into the atmosphere (Bessa et al., 2020). Several studied ceramic factories have an extraordinary potential for airborne UFPs materialization and discharge to the worked area. Fire and burning procedures are also related with diffusion of wood NPs in the atmosphere, indicating a large hazard (NIOSH, 2009). For example, NPs comprising Al-, Cd-, Cr-, and Cu-oxides have been linked with ceramics industry (Bessa et al., 2020).

Alumina, ceria-NP, synthetic organic compounds (i.e. graphene, fullerenes, carbon nanutobes), hazardous elements (i.e. Sb, Cd, Cu, and Pb), sulfides (e.g. ytterbium), and oxides (e.g. ZrO_2, NiO, ZnO, and Cr_2O_3) are used in several modern ceramics to improve stiffening fillers in order to make cutting tools (Bessa et al., 2020). In the present study, as it is in an area of handmade ceramics, such advanced materials were not detected. It should be noted that this is one of the few advantages of artisanal ceramic production, since such NPs have been reported by many studies as harmful to health (Akinyemi et al., 2020; Oliveira et al., 2019a,b,c; Saikia et al., 2014). On the other hand, the lack of proper filters in the chimneys shows an enormous potential for exposure to workers and to residents in the proximity of the ceramic factories under study.

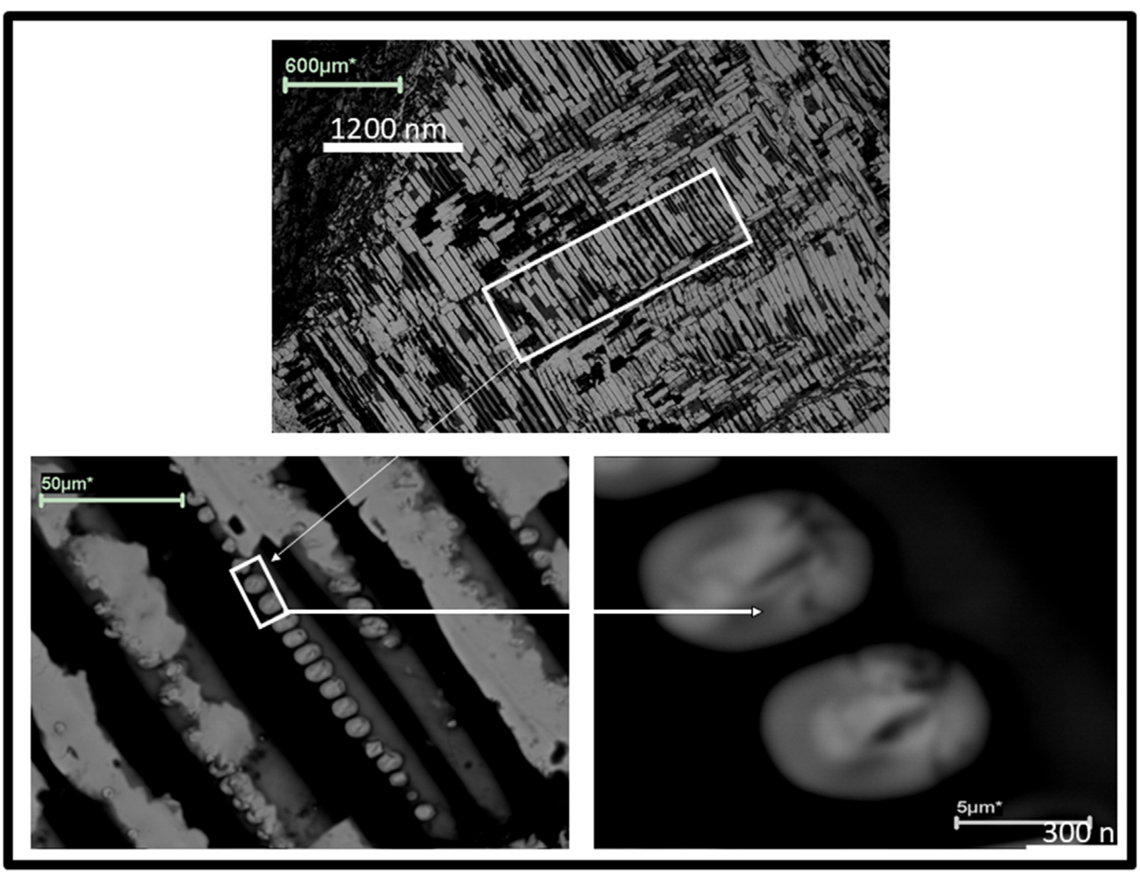

Fig. 4. Typical pyrite grains detected by FE-SEM and analyzed by FIB-SEM.

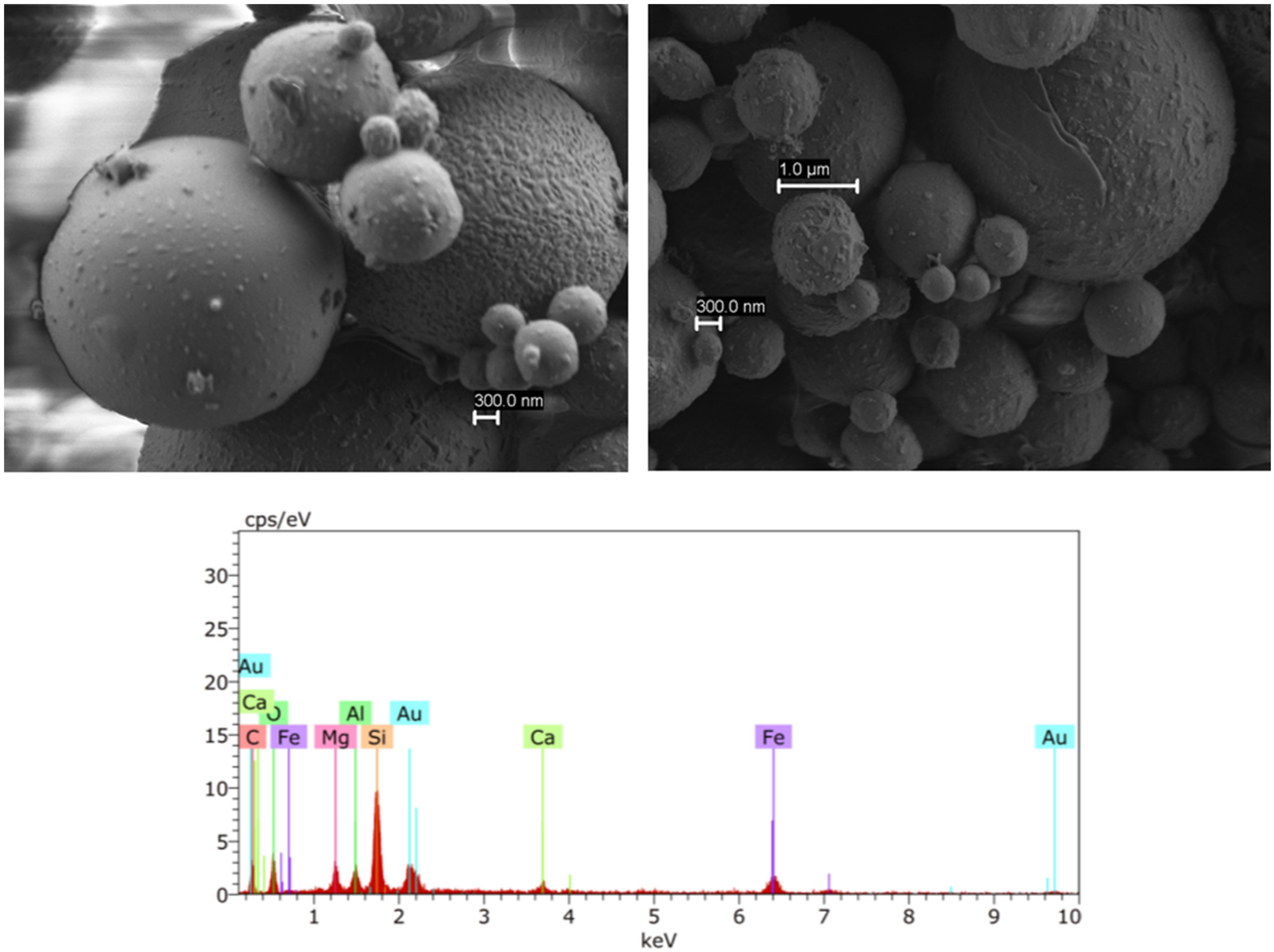

Fig. 5. Illustrations of probable ultra-fine coal fly ashes and general EDS (Au peak from samples coated with gold).

Organic and inorganic nanocompounds and mixtures of both nanocompounds originate in ceramic factories and impacting contact are normally created from two bases: (1) discharge resultant from industrial actions and (2) circumstantial particulate matter. In NPs and UFPs from the studied area, these result from artisanal activities (e.g., wood combustion) and from novel NPs creation (e.g., ceramic production and air nucleation). Similar results were previously reported by Salmatonidis et al. (2018) and Bessa et al. (2020), among others.

The most abundant particles are clays and quartz, typically soil material, and particulate matter. In general, inorganic particles larger than 100 nm did not contain toxic elements. The <100-nm particles contained elements such as Ti as a constituent of rutile and anatase (both detected in this study) to some Fe hydr/oxides contained minor portions of Cr and Se. Although the use of clays with a high proportion of organic matter can be observed visually in the ceramics production under study, it is believed that this factor, the use of wood combustion is the major generator of organic particulate matter. This is because organic-rich clays are rarely used according to ceramic manufacturers, while wood combustion is always used.

Clays were the most common compounds utilized in the studied ceramic production There has been an extensive application of nanosized aluminumsilicate in the factory, which increases concerns for the possible dangers of NPs for the unprotected personnel health. For example, Wagner et al. (2018) demonstrated that nano-clay contact reduces cell capability and encourages modifications in morphology. The acquired results showed that these materials provoked irritation and granuloma materialization (Stueckle et al., 2018; Oliveira et al., 2021).

The wood-combustion particles were easily distinguished thanks to the EDS analyses that proved a high proportion of K in the carbonaceous matter (Fig. 6). According to previous authors, wood-combustion ash contains high concentrations of these elements and others, including Si, P, and Ca (Vicente et al., 2020). The variances in the wood-burning working circumstances were manifested in diverse UFPs geochemical compounds and, subsequently, in different human dangers. The hazard level in this work depends on a contact situation instead of the characteristic danger quantification established on permanent contact.

4.3. Complex mixtures

Fig. 7 illustrates the diverse shapes, morphologies, sizes, and complex compositions of the main NPs detected in this study. In general, we can observe that it is difficult to resolve the crystalline structure because the NPs are, in most cases, agglomerated with carbonaceous phases, making the study difficult using FFT and SAED. Recent studies have shown that it is possible to detect the crystalline structure of some NPs more easily, especially since they did not occur as agglomerates with organic phases (Kronbauer et al., 2013; Ribeiro et al., 2013a; Martinello et al., 2014; Oliveira et al., 2019b). This is probably since, in coal-fired power plants, the organic matter is largely transformed into energy and some contaminants in the ashes, there is little concentration

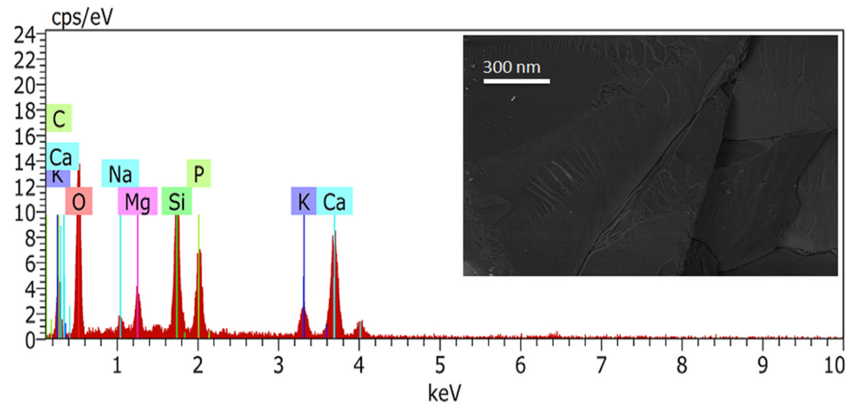

Fig. 6. Classical organic material concentrated on elements typically detected in wood combustion ashes.

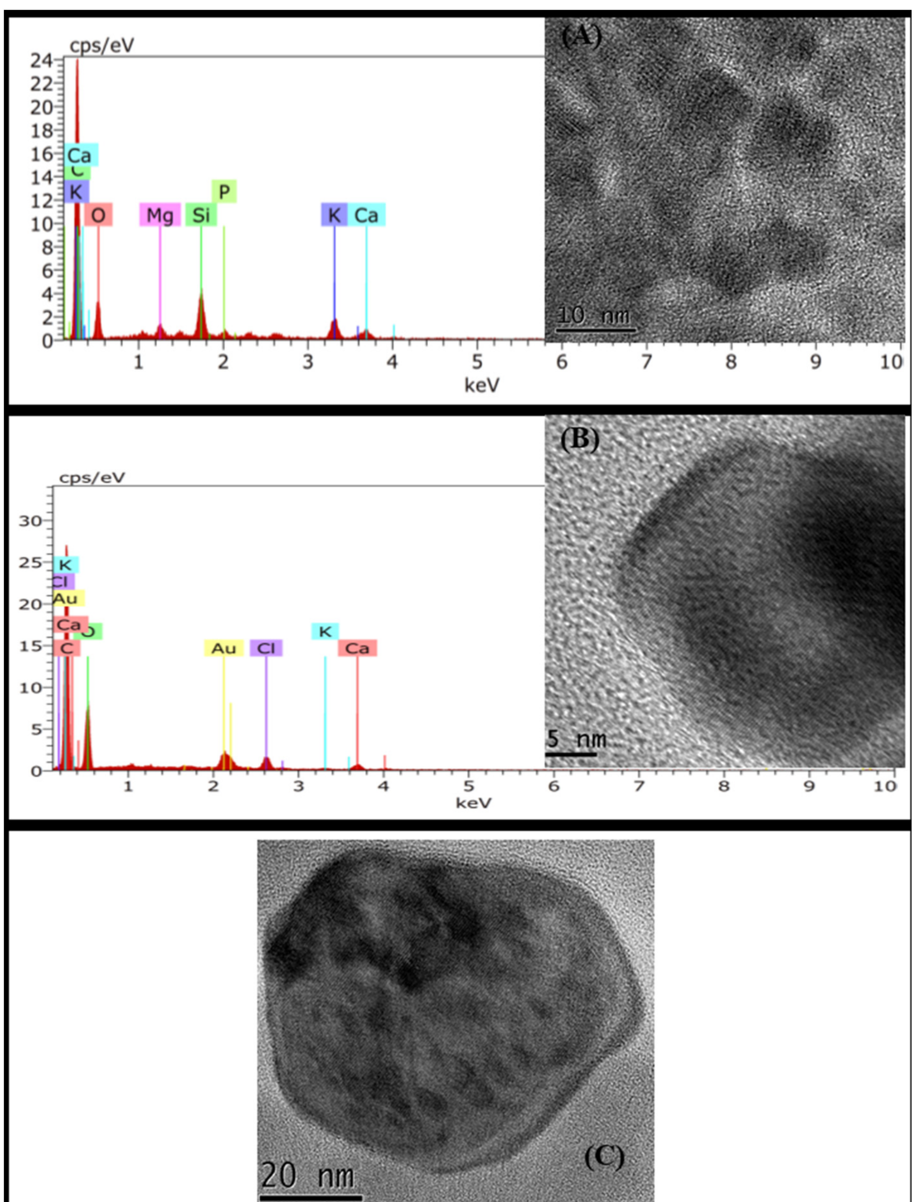

Fig. 7. Principal NPs detected. (A and B) Carbonaceous complex particles and (C) Rutile. Au peak from samples coated with gold.

of carbonaceous matter (Silva et al., 2009; Wilcox et al., 2015). Fig. 7A shows the greater tendency for particles to contain residues of wood combustion, since according to some previous authors, K, Ca, and P are tracers of wood combustion (Vicente et al., 2020). There is a significant concentration of elements such as Cl (Fig. 7B), which are usually tracers of sea- (because the factories are close enough to ocean) and coal-derived particles, according to previous studies of the mineralogy of the Santa Catarina coals (Oliveira et al., 2013). Rutile (Fig. 7C) is one of the few nanominerals that could be detected in the sampled samples. This mineral can occur in both soil and coal ashes, so it is difficult to define the exact origin. Several toxicological studies report that it is a NP that can generate oxidative stress, cell damage, genetic mutations, and several other negative health effects (Oliveira et al., 2017; Yang et al., 2017; Lütke et al., 2020; Silva et al., 2020b). Potentially hazardous elements typically exist at low levels in woods, but after the burning they persist in the wood ash at moderately high concentrations (Nzihou and Stanmore, 2013).

Considering that the coal-fired power plant and ceramic companies, the number of vehicles, especially buses and diesel trucks, have been growing; consequently, the population of this study area, being located close to highways, is exposed to a complex mixture of organometalic compounds (Ramírez et al., 2019, 2020; Rojas et al., 2019; Silva et al., 2020a). Future studies will be necessary to prove the real dose-response of the exposure of workers in artisanal ceramics. After all, as noted, countless organic, inorganic, and complex mixtures are formed due to the handling of clays, the wood combustion, and the lack of personal protection equipment. Only with mechanized industrialization and more active legislation will it be possible to reduce the degree of nanoparticle exposure of people who work and live near ceramic's plants in southern Santa Catarina.

5. Conclusions

Despite their importance to the local economy, artisanal ceramics have a serious impact on the environment and on the health of workers and the population. Local authorities financially support the mechanization of such factories, as well as enforce environmental laws that protect the environment, the health of workers, and the local population. The results of this study demonstrate a wide variety of particles both in terms of size, morphology, and composition. Source estimation must also be applied to properly sanction the region's industries, as well as to identify specific toxicological evidence. In order to predict whether the ultra-fine and nanometric spheres of coal fly ashes are more or less toxic than the UFPs and NPs resulting from ceramic activities, toxicological studies that directly compare coal fly ashes, wood combustion ashes, and residual ceramic dust are necessary.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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