Article

Worldwide Survey and Characterization of Ultrafine Particle Exposure Monitoring and Assessment

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Abstract: Several studies show that air pollution has become a major problem and a source of great concern. Part of the observed poor air quality can be attributed to the levels of ultrafine particles (UFP), generally defined as the fraction of particles with diameter less than 100 nm which, by their size, are able to enter the blood circulation causing adverse health effects. As the main route of entry into the body is inhalation, several epidemiological studies associate the presence of UFP to total mortality, cardiovascular and respiratory problems. Since there are apparently no worldwide normative references for the control of these particles, this global study aimed to characterise the methods and techniques used worldwide regarding the monitoring and management of UFP. For this purpose, a survey was carried out based on a questionnaire applied at global scale. The questionnaire design was based on a preliminary literature review on the main search engines for scientific publications to establish the state of the art. As a result of the application of the questionnaire, a total of 51 responses were obtained from 20 different countries on five continents, which provided an insight into the global profile of professionals interested in the control/study of UFP, the measurement methodologies they follow and the guideline values they apply, if any. Among them are public health professionals, academics and health engineers from various institutions, such as education, hospitals, research centers and regulators in the health sector. Most responses said are no specific methodologies for the measurement of UFP, nor national standards for it. Through the results obtained in this study, it can be concluded that it is urgent to adopt regulations to standardize the assessments as well as to establish guide values for the protection of the human health.

Keywords: Ultrafine Particles 1; Air Quality 2; Occupational Health 3; UFP worldwide survey 4

1. Introduction

Particulate matter (PM) is a mixture of solid or liquid airborne particles of different sizes and compositions containing a variety of constituents such as dust, dirt, soot, pollen or small metal and plastic particles, smoke and liquid droplets, some of which may be toxic and classified as pollutant. These particles are PM_{10} (i.e. particles up to 10μm in aerodynamic equivalent diameter), coarse particles or $PM_{2.5\text{-}10}$ (particles between 2.5 and 10μm in aerodynamic equivalent diameter), $PM_{2.5}$ or fine particles (particles up to 2.5μm in aerodynamic equivalent diameter) [1].

Particle size is important because it affects how aerosols interact with their environment. Smaller particles can stay in the air longer, travel further and penetrate deeper into the lungs. Larger particles, on the other hand, are more likely to fall out of the atmosphere quickly and are less likely to reach the lungs. In addition, the size of particles can affect how they interact with other components of the environment, such as water vapor, sunlight and other pollutants. Particle size can also affect how

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aerosols interact with other particles, such as how they agglomerate to form larger particles. All of these factors can have a significant impact on air quality and public health [2].

The term "equivalent diameter" describes the size of particles of unknown composition and/or shape as spheres of specified density. UFPs are generally considered to be particles with a diameter of 100 nm or less. There is already considerable evidence of the toxicological effects of UFP. However, at an early stage, evidence from epidemiological studies was insufficient to establish guidelines, although now an increasing number of studies does support the need to establish reference guidelines [3].

Over the last 30 years, $PM_{2.5}$ and PM_{10} have been the most used parameters to assess human exposure to particulate matter. Either $PM_{2.5}$ or PM_{10} also includes the fraction below 100 nm (i.e. ultrafine particles) but, in fact, UFP is a minor contributor to the total mass concentration of indoor and outdoor aerosols [2]. Because of that, the UFP concentration is define as the particle number concentration (i.e. the number of particles in a given volume of air), as the mass concentration is too low to be measured reliably and effectively. There are two main aspects that separate UFP from larger airborne particles ($PM_{2.5}$/PM_{10}) in terms of toxicology: (i) differences in the deposition (local dose) upon inhalation, and (ii) differences in intrinsic toxicity as a result of physicochemical properties. Of note, associations have been observed between UFP exposure and health effects independently of other metrics of air pollution such as $PM_{2.5}$ and NO_x. Interestingly, some UFP might translocate out of the lung and then reach the circulatory system and other organs, while this is not the case for larger PM [4].

One of the most important anthropogenic sources of UFP is combustion. This includes primary emissions from industrial plants, vehicles and machines that use biomass or fossil fuels. Vehicle emissions, including non-exhaust emissions, are a major source of UFP in urban areas [5]. Other sources of UFP are the secondary formation who is a complex and dynamic process influenced by several factors, including precursor gas concentrations, atmospheric conditions and the presence of other aerosol particles. This process occurs when the concentration of precursor gases reaches a critical level and favorable environmental conditions, such as low temperature and high humidity, are present [6].On the other hand, looking to buildings air quality, any type of indoor activity can produce a large amount of UFP [7]. Several studies have identified sources of UFP in dwelling indoor air from activities such as cooking, smoking, burning candles, cleaning, using a spray, ironing, electric heating, vacuum cleaning and printing [2,8,9].

The deposition patterns and clearance mechanisms of aerosol particles depend mainly on particle size. In general, larger particles (> 10 μm) tend to be deposited in the nose and upper airways, whereas smaller particles (< 10 μm) are more likely to reach the alveolar region. The clearance mechanism for larger particles is mainly due to sedimentation, whereas for smaller particles it is mainly due to the action of alveolar macrophages. In addition, deposition patterns and clearance mechanisms are influenced by particle shape, particle density and the breathing patterns of the exposed individual [10].

The first site of interaction for $PM_{0.1}$ is the lung. The surface area of the lung is estimated to be greater than 100 m^2, but this is usually estimated by measuring linear intersections with a 1 μm probe by light microscopy. The estimated surface area will be greater the smaller the sampling probe is, to account for the additional area of an irregular surface. The surface area of the lung for a $PM_{0.1}$ nanoprobe would be orders of magnitude larger than the light microscopy estimates, which Weibel called the "Coast of Wales" effect [11].

Studies have shown that UFP causes systemic inflammation and coagulation changes that predispose to ischemic cardiovascular disease, circulating polymorphonuclear leukocytes, platelets, fibrinogen, plasma viscosity and other markers. UFP promotes endothelial dysfunction, vascular inflammation, and atherosclerosis. Previous studies have attributed this effect primarily to $PM_{2.5}$, but a growing number of studies show that UFP plays an important role in virtually all of these factors. In fact, most studies show a much broader effect, as UFP also causes increased heart rate variability, loss of sympathetic vagal balance and altered inflammatory and hemostatic functions in exposed humans [11].

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There published work on the effects of PM on the brain or nerves, as well as on the mechanisms by which UFP affects the brain and its development. Translocated UFP can be found in the brain after inhalation. UFP inhaled through the nose can reach the brain via the olfactory nerves. After exposure to UFP aerosols, brain uptake is greatest in the olfactory bulb, even 7 days after exposure. In an animal inhalation study, up to 20% of UFP deposited on the olfactory mucosa reached the olfactory bulb. This route, which may bypass the blood-brain barrier, may be even more direct in humans. UFP not only translocate and directly damages nervous tissue, but also affects autonomic function. Exposure to UFP increases sympathetic nervous system activity by decreasing norepinephrine clearance, a property that is enhanced in humans [12].

Assessing exposure to UFP is much more complex than assessing particulate matter ($PM_{2.5}$ or PM_{10}) because UFP concentrations exhibit considerable spatial and temporal variations within a few seconds and over a few meters, which can be up to an order of magnitude above background levels as people move closer to or away from pollution sources or move between different microenvironments [13]. These large variations in UFP concentrations in different environments may be important for human exposure assessment and epidemiological studies [14]. The influence of temporal activity and movement can easily be overlooked when using averaged results, so that mean and median concentrations over a time-averaged period may not reflect all aspects of population exposure patterns. It should be kept in mind that an exposure assessment approach for epidemiological studies should be designed to be easy to measure and universally applicable so that uncertainties shrink, but still be able to relate it to entities that can be measured, modelled and also regulated. The absence of an exposure-response relationship makes it difficult to propose health guidelines for UFP, so it is not surprising that, to the best of the authors' knowledge, there are still no air quality guidelines for UFP anywhere in the world [2].

However, the European Commission has already set out its intentions to monitoring and control the UFPs levels regarding indoor air quality in the Directive of the European Parliament and of the Council on ambient air quality and cleaner air for Europe, published on 26 October 2022 [15] and WHO has also expressed concern about UFP, suggesting distinguish between low (< 1000 particles.cm^{-3}, 24 hour mean) and high Particle Number Concentration (PNC) (>10 000 particles.cm^{-3}, 24 hour mean or 20 000 particles.cm^{-3}, 1 hour) to guide decisions on priorities for emission control of UFP sources [3]. Safe Work Australia and the British Standard Institute have also provided some references and concern to UFP control [16].

On the other hand, the literature review showed that only the ISO 16000-34:2018 give us a method for all the Particulate matter (PM) measurements, including UFP.

There seems to be a huge information gap on UFP on such common aspects as who, when and why monitors this pollutant, what means and methods they use and, finally, whether there is a guideline reference value.

In this context, this work had the following objectives:

- Are there national laws or regulations that set guideline values for UFP?
- What methodologies and instruments are used by the international community to assess UFP?
- Who monitors and assesses UFP exposure?

To answer these questions, a literature review on UFP monitoring was firstly conducted, assessing the possible existence of limit values as well as the existence of regulations and standards for monitoring methods. Afterwards, a worldwide survey based on a questionnaire was distributed to the international scientific community. The results of that questionnaire are interpreted and discussed in this manuscript.

2. Materials and Methods

In order to elaborate the questionnaire presented in this work, a bibliographic research was carried out in the scope of UFP, which allowed the identification of the measuring methods generally used and their gaps, as well as the knowledge of the existence, or not, of regulations in this subject.

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Subsequently, a questionnaire was created in English using Google forms. In order to test the level of understanding and intent of the survey, ensuring its reliability, a pilot questionnaire was carried out and presented to three native English speakers and five native speakers of other languages. The pilot test included fields for comments, which were carefully analyzed and taken into account in the review of the structure and functioning of the questionnaire. After the evaluation, the questionnaire was revised and sent through International Federation of Environmental Health (IFEH), Sociedade Portuguesa de Saúde Ambiental (SPSA) and other international contacts.

In the first section of the questionnaire a sociodemographic characterization of the responder was create, with close questions asking for age, gender, and main area of professional activity and main área.. In a second part of the questionnaire, several questions were asked about how UFP are controlled in their countries of origin. For example, it was asked if assessment of environmental exposure to UFP is common in their countries or if assessment of occupational exposure to this pollutant is also common. They were also asked whether they were aware of the existence of national legislation for exposure to UFP in both environmental and occupational fields, and whether there are standards for measuring them. At the end of the questionnaire was asked about the time, frequency and equipment used to the UFP assessment. After coding the questionnaire, it was processed and statistically treated using IBM SPSS 25.

3. Results Presentation and Discussion

The questionnaire was applied to 250 entities in 20 countries resulting in 51 valid responses from 16 countries. The results allowed to verify which professions/training courses control/study the ultrafine particles, as well as the objectives and the means used (equipment and measurement methodologies applied).

The majority of respondents stated that there are no specific methods for dealing with ultrafine particles, nor are there any national standards for them.

Table 1 shows the characterization of participants by age and gender.

Table 1. Characterization of participants by age and gender.

		Occurrence	Frequency (%)
Age	<25	6	11.8
	26-35	7	13.7
	36-45	16	31.4
	46-55	15	29.4
	>56	7	13.7
	>56	7	13.7
	Total	51	100.0
Gender	Female	28	54.9
	Male	23	45.1
	Total	51	100.0

It can be seen that the highest frequency of occurrences is in the age group 36-45 and 46-55 years, with 31.4% and 29.4% respectively. On the other hand, about 26% of the frequency is below 36 years and about 14% above 56 years.

With regard to gender, the distribution of technicians involved in this type of measurement is relatively balanced, with around 54% women and 46% men.

Concerning the participants' countries of origin, responses were obtained from 16 countries on five continents (Table 2).

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Table 2. Countries of origin of the questionnaire participants.

Countries	Occurrence	Frequency (%)
Australia	6	11.8
Belgium	1	2.0
Bosnia and Herzegovina	1	2.0
Brazil	3	5.9
Canada	3	5.9
China	3	5.9
Côte d'Ivoire	1	2.0
Croatia	10	19.6
Estonia	2	3.9
France	4	7.8
Germany	1	2.0
Portugal	2	3.9
Slovenia	8	15.7
Spain	3	5.9
USA	2	3.9
Zimbabwe	1	2.0
Total	51	100.0

Over 60% are Europeans, about 16% are from American countries (which include Canada, Brazil and USA), 12% from Australia, 6% from China and 4% are from Africa. The country with the largest participation was Croatia (10 participants) and three countries had only one participation each (Belgium, Bosnia and Herzegovina and Zimbabwe).

Table 3 shows the professional affiliation of the survey participants.

Table 3. Professional affiliation of the survey participants.

	Occurrence	Frequency (%)
Academia and Research	14	27.5
Biomedicine	3	5.9
Environmental health	22	43.1
Microbiology	2	3.9
Occupational safety	4	7.8
Researcher	3	5.9
Total	48	94.1
No answer	3	5.9
Total	51	100.0

As can be seen, the largest proportion, almost half (43.1%), is from the environment and health sector, followed by the academic and research sector (27.5%). The remaining participants are from the occupational safety, research, biomedicine and medicine sector.

Concerning to participants workplace, they mainly work at education sector (41.2%) and regulators, with about 20% of the participants (Table 4). The remaining participants work in research centers and other locations, which may indicate that UFP is being assessed, not for research, but in routine measurements in worker protection.

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Table 4. Participants professional workplace.

	Occurrence	Frequency (%)
Education Institution (non-hospital)	21	41.2
Education Institution (non-hospital), Research Centre	3	5.9
Hospital or Teaching Hospital	3	5.9
Regulator	10	19.6
Research Centre	9	17.6
Other	5	9.8
Total	51	100.0

These results are concordant with the need to systematically and quantitatively assess the existing evidence based on epidemiological research. These analyses should consider the heterogeneity of source contribution patterns for UFP in different regions with climatic and emission patterns [4].

Table 5 shows the results by professional work area of the participants.

Table 5. Participants professional work area.

	Occurrence	Frequency (%)
Occupational Health	4	7.8
Environmental	9	17.6
Public Health	11	21.6
Environmental Health	9	17.6
Public Health and Environmental	10	19.6
Other	8	15.7
Total	51	100.0

There is a large convergence in the fields of work of the respondents. This figure is slightly lower for occupational health workers. These figures are similar to what would be expected, as the vast majority of the studies found are in the field of indoor and outdoor air quality, which is actually studied by public health and environmental health [5,17].

Participants were also asked if they had experience in measuring UFP (Table 6).

Table 6. Participants experience in UFP measurement.

With experience	Occurrence	Frequency (%)
No	19	37.3 %
Yes	15	29.4%
Yes, some	17	33.3%
Total	51	100.0%

More often, 37.3%, stated that they had no experience of measuring UFP, while 33.3% reported having some experience. A slightly smaller proportion (29.4%) reported having some experience. As a new area of intervention, these values are not surprising and are in line with studies that point to the lack of studies and the need for more and better studies in different countries worldwide [1,3,13].

When participants were asked whether they knew if UFP monitoring is carried out in their country for IAQ, it was found that, 70.6% said yes, 25.5% said they did not know and only 3.9% said no (Table 7).

Regarding occupational exposure, in Table 7 can be observed that there is less reference to measurements being taken, with only 49% of the responders saying yes. A very similar number, 43.1%, say they do not know and an insignificant number, 7.8%, say that this type of measurement is not carried out..

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From the literature review, studies and assessments of UFP are in very specific areas, such as during laser hair removal procedures [18], steel industry, police officers [19] and taxi drivers [20]. or occupational exposure to particulate UFP in metal additive manufacturing [21] However, survey participants have a higher number of responses referring to the assessment of indoor air quality in buildings. We consider that this may be justified by the growing interest in this research area.

Table 7. UFP monitoring objectives in the collected responses.

		Occurrence	Frequency (%)
Indoor air quality	No	2	3.9
	Yes	36	70.6
	I don't know	13	25.5
	Total	51	100.0
Occupational (indoor)	No	4	7.8
	Yes	25	49.0
	I don't know	22	43.1
	Total	51	100.0

From all the answers to the questions asked on this matter, one gets the idea of uncertainty on the part of those who are making the UFP measurements about the reason/objective for the measurement.

When we asked the participants about the existence of standards and legislation in the control and monitoring of UFPs, some mentioned their existence (Table 8).

Table 8. National Legislation and standards in UFP.

		Occurrence	Frequency (%)
Indoor air quality	No	12	23.5
	Yes	23	45.1
	I don't know	16	31.4
	Total	51	100.0
Occupational (indoor)	No	8	15.7
	Yes	25	49.0
	I don't know	18	35.3
	Total	51	100.0

In fact, 45.1% of respondents for IAQ and 49% for occupational assessments, confirmed the existence of legislation or legal standards in their country. Only 23.5% of respondents for IAQ and 15.7% for occupational assessments, respectively, stated that there was no standard or legislation applicable to UFP in their country. It should be noted that about 1/3 of the respondents did not know whether or not there were standards or legislation in their country. These figures contradict the literature review conducted in this work where no references to legal standards for the assessment or control of UFP were identified, either for indoor air quality or occupational health control. Nevertheless, this concern has already led the World Health Organization to lunch "global air quality guidelines" [3].

However, Safe Work Australia proposes to include standards aggregates and agglomerates with a size >100nm, leaving the threshold undefined. In this guidelines UFPs are categorized into different risk groups, which may vary from organization to organization. In the British Standard Institute's approach, four groups provide a basis for categorizing nanomaterials, but without defined values [16]. In fact, workers are exposed to UFPs in a variety of work environments, but this exposure is not currently regulated as a separate part of the usual occupational exposure limits [22].

On the other hand, as mentioned earlier, concern about UFP has already led the Guideline Development Group from World Health Organization to establish best practice limits [3] the

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European Parliament and the Council to propose an obligation to measure UFP in their Directive on ambient air quality and cleaner air for Europe, published on 26 October 2022 [15].

From these responses, we begin to realize that there may be some confusion between measurements of UFPs and those of IAQ or occupational exposure to inhalable and respirable particulates. Particularly only in recent years, emerging industry has drawn attention to worker exposure to ultrafine particles. These have been measured in several workplace studies and several review articles have been written on the subject [22,23].

When asked whether they use a legally established methodology for UFP measurements, only 23.5% said yes and 76.5% answered no (Table 9).

Table 9. Legally established methodology for UFP measurements.

	Occurrence	Frequency (%)
No	39	76.5
Yes	12	23.5
Total	51	100.0

When the participants were asked about the duration of the measurements and the routine of the procedures, we obtained some relevant results. Of the 51 participants, only twelve (24%) mentioned the existence of a routine in the measurements. The vast majority (76.5%), did not know or did not answer on this subject. Given these data, apparently UFP is not routinely measured for occupational health or for indoor air quality control purposes. In addition, measurement methods and instruments are not currently standardized, which means that measurement strategies and methods vary widely. Some studies have sampled workers' in breathing zones, while others have used stationary sampling. The sampling distance has a significant influence on the measurement results, as the concentration of UFP is rapidly diluted after leaving the original exposure source [22]. Table 10 shows the Measure time and routine in UFP assessment.

Table 10. Measure time and routine.

		Occurrence	Frequency (%)
Measure Time	1 min	2	3.9
	5 to 10 min	6	11.8
	more than 15 min	4	7.8
	doesn't know or doesn't answer	39	76.5
Measure routine (number of measures made in each point)	one	3	5.9
	three	3	5.9
	More than three	6	11.8
	doesn't know or doesn't answer	39	76.5

Considering the equipment mentioned by the participants for the quantification of UFPs we checked with the manufacturers' websites for their characteristics.

Laser Particle Sensor - PM2008-API, is a laser particle sensor module, based on laser scattering technology ultra-thin (12mm) designed with all metal shielding. Detecting particle concentration size between 0.3 μm – 10 μm in the air and real-time output $PM_{1.0}$, $PM_{2.5}$, PM_{10} in $\mu g.m^{-3}$ via mathematical algorithm and scientific calibration.

Fluke 985 Particle Counter – Is a Particles laser couter to sizes 0.3μm, 0.5μm, 1.0μm, 2.0μm, 5.0μm, 10.0μm. This equipment is not valid for UFP measurement.

GRIMM Aerosol has been standing for the optical aerosol measurement made in Europe. The measuring range of our systems extends from less than 1 nanometer to 35 micrometers. The measuring instruments are usable stand-alone or integrated in measuring containers. The analysis is made via an intelligent evaluation and control software.

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TSI The P-Trak Ultrafine Particle Counter TSI The P-Trak Ultrafine Particle Counter detects and count Ultrafine particles. The optical counter gives direct real-time measurement of workplace ultrafine particulate levels.

Table 11 shows the used equipment by the responders of the survey.

Table 11. Used equipment by the responders of the survey.

	Occurrence	Frequency (%)
GRIMM	2	3.9
Laser Particle Sensor PM2008-API PM2008-API	5	9.8
FLUKE 985 particle counter	2	3.9
TSI counter	4	7.8
not known	1	2.0
Total	14	27.5

It was possible to verify the variability of the equipment used, some accurate and advanced, such as the Laser Particle Sensor PM 2008-API, whose cutting-edge technology allows the sample characterization by size (5 particles) or the TSI UFPs Counter, specifically designed for occupational hygiene (4 participants), or the Grimwhich later allows an evaluation with quantification in the laboratory. However, two of the participants reported using the FLUKE 985, designed for $PM_{2.5}$ and PM_{10} assessment and not applicable to UFP measurement. We believe that this confusion may be related to the fact that there is no specific regulation or legislation defining UFPs.

4. Conclusions

The added value of the application of this survey is a global knowledge of how are dealing with UFP. In fact, information was obtained from 20 different countries in five continents making this survey, to the best of the authors' knowledge, the most extensive work done on this subject.

The following conclusions could be drawn:

- The inexistence of guideline values for the assessment of UFPs. To the best of the research team's knowledge, monitoring and quantifying ultrafine particles (UFP) is not yet required by law in any country; only WHO and the European Commission show some concern about this pollutant.
- There are no defined protocols for the assessment of UFP levels, which means that assessments do not follow the same methodologies, making it impossible or very difficult to intercompare the results obtained; The literature review showed that only the ISO 16000-34:2018 give us a method for all the Particulate matter (PM) measurements, including UFP;
- Lack of knowledge about UFP; Some of the respondents confuse ultrafine particles with the rest of the particles; this is evident by the type of equipment they say they use to measure UFP which, according to the technical characteristics, is not suitable for the purpose.

International methodologies for the measurement of UFP are urgently needed, as well as the establishment of guide values for the protection of human health. This need is reflected by the respondents, most of whom are academic heads. They are concerned about the emerging issue and the need to control and set standards to monitor this pollutant. Until then, we suggest using the ISO standards (ISO 16000-34:2018) to determine the level of UFP and the best practice limits already advanced by WHO.

Author Contributions: F.M, A.F. and N.B. designed the study. F.M. Design the questionnaire, collected the data and performed the statistical analysis. F. M., A.F. and N.B. prepared the original draft of the manuscript. F. M., A.F. and N.B. critically interpreted the results, reviewed the draft version, and approved the final manuscript. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement: To carry out the study, authorization was obtained from the President of the Higher Education Institution evaluated. The study protocol was approved by the IES data protection officer.

Informed Consent Statement: Informed consent was obtained from all the subjects involved in the study.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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