

## SOURCE APPORTIONMENT OF OXIDATIVE POTENTIAL What We Know So Far

by

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*In numerous epidemiological studies, exposure to particulate matter (PM) has been associated with negative health outcomes. It has been established so far that the detrimental health effects of particles cannot be explained by a single parameter, such as particle mass, as the complexity of chemical composition and reactivity of particles are not always represented by the mass loadings. The oxidative potential (OP) of aerosol particles represents a promising indicator of their potential toxicity. To develop strategies and regulations at improving the air quality, an increasing number of studies are focused on the application of source apportionment (SA) of PM., while a limited number of SA investigations have been applied to OP. In this review previous research of SA of atmospheric PM OP and proposed guidelines for future studies are summarized. Most of the research studies were carried out in an urban area and focused on PM<sub>2.5</sub>, while few studies examined other PM fractions. It was noted that the three dominant contributors to OP were biomass burning (9-97%), secondary aerosols (6-67%), and traffic/vehicles (16-88%). The presence of other factors that contributed to the increase of OP to a lesser extent depended on the location and season. Further, a considerable discrepancy in the contribution of various OP vs. PM sources was discovered using SA models. Because of this, the use of SA is not equivalent when considering the mass of PM and its toxicity.*

Key words: oxidative potential, source apportionment, toxicity

### Introduction

The fast changes in pollutant emissions brought on by comprehensive human activity and rapid industry development make air quality one of the main issues in the urban environment. The effects of air pollution on visibility, human health, and global climate change have generated considerable public concern [1, 2]. As a result, quantifying different pollutants

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in the air has become crucial for authorities to analyze air quality and enact regulations that will help reduce pollution. Although most cities worldwide have policies to control certain pollutants, in urban environments, severe haze episodes still occur primarily during the winter heating season. Also, natural fires that occur during the summer period, when they are difficult to control due to unfavorable weather conditions, also greatly impact air pollution. Numerous research has suggested that PM is one of the most important pollutant influencing air quality and affecting the greatest number of people. Hence, it is essential to have accurate PM monitoring to create services to lower the amount of pollutants that people would be inhale.

The complexity of PM comes from various factors: the varying chemical composition, size of particles, meteorological conditions, emission sources, a great number of transformation reactions, aging in the atmosphere, different measurement, and detection techniques. The PM mass concentration has been employed as an exposure indicator, however, it may underestimate PM's total impact since it neglects the sizes, composition, toxicological effects, and interactions of its components. Furthermore, a single metric, such as particle mass concentration, cannot fully explain the negative health impacts of particles, and as a consequence, epidemiological study results are fraught with ambiguity. Major chemical components (*i.e.*, mass contributors) may have low or extremely low toxicological activity, causing modest or no detrimental impacts on health, while trace chemical components may have significant toxicological activity. In this regard, PM mass concentration is an inadequate measure for describing the mechanisms through which PM exposure might affect human health. This constraint can be eliminated by determining whether or not there is a connection between PM toxicity and the particular physical and chemical features of PM. In recent years, the PM composition has been the subject of numerous researches. Several PM components have been recognized as crucial in determining its possible health and environmental impacts.

Although there are several theories on how short-term or long-term exposure to particles can affect human health, one of the leading theories suggests that the detrimental effect of PM can be attributed to its oxidative or oxidant-generating properties. According to current studies, many negative health impacts are induced by oxidative stress in biological systems produced by PM accumulation in the lungs. Oxidative stress is the imbalance that results from increased radical generation and a decreased capacity for the body to deal with excess radicals or repair the damage caused by radicals [3]. The oxidative stress paradigm is the name given to this developing theory, which states that oxidative stress might occur as a result of two different mechanisms. The first mechanism is associated with the intrinsic oxidation-reduction processes caused by the redox-active compounds contained in PM. These reactions are caused by exogenous reactive oxygen species (ROS). The ROS is the collective term for chemically reactive oxygen radicals, such as: superoxide ( $O_2^-$ ), hydroxyl (OH), peroxy ( $RO_2$ ), and alkoxy (RO), as well as oxygen-derived species, such as: hypochlorous acid (HOCl), ozone ( $O_3$ ), singlet oxygen ( $^1O_2$ ), and hydrogen peroxide ( $H_2O_2$ ), all of which are oxidizing agents [4]. The second mechanism is the biological reaction to inhaled PM or cell-mediated oxidant-producing capacity. Because of this, a thorough investigation of how airborne PM affects human health is required.

### Reactive oxygen species

Atmospheric ROS can be introduced into the body by inhaling particles that already have ROS attached to them (particle-bound ROS). The ROS can be generated through various *in vivo* reactions catalyzed by specific components present in the particles, defined as OP [5, 6]. Since the direct measurement of ROS in the air is a complex process, in order to assess the

toxicity of particles, the determination of their OP is increasingly being used. The OP is defined as a measure of the ability of particles to oxidize certain molecules, resulting in the formation of ROS [7]. Thus, oxidative stress is a key PM toxicity mechanism, and measuring PM OP might help elucidate downstream pathways. However, monitoring the ROS parameter in the environment may be challenging due to the complex measuring methods involved in biological systems. Also, ROS may be found in the gas phase as well as the particulate phase. Further, there are two types of ROS, short- and long-lived ROS. The former has a half-life of only a few minutes, while the latter can be found in the air for several hours to days [8].

In current research, the measurement of OP is performed using various cell-free tests (acellular assays), as well as *in vitro* and *in vivo* tests. Consumption of a different chemical (often antioxidants) or direct detection of ROS production from PM extracts constitutes the basis of cell-free assays. Acellular assays have limited physiological validity, as they are performed without the actual interaction that occurs between the cell and the particles. On the other hand, these assays are easier to use than cell trials and allow relatively fast data reading. They are helpful for a rapid initial hazard screening and require less controlled environments. The advantage of these tests in relation to *in vitro* measurements is reflected mostly in the lower cost of reagents required for analysis, as well as the shorter time needed for the application of the assay.

Both *in vitro* and *in vivo* methodological approaches aim to investigate the biochemical and molecular mechanisms of toxic particles. In addition, these tests provide insight into the specific negative effects that particles can cause in humans. *In vitro* methods are based on cultured cells that can be immobilized or freshly collected. Lung cells are most commonly used to analyze ambient particle toxicity [9-11]. These measurements can detect specific changes that occur on the surface or inside cells, which helps to better understand the mechanisms of particle toxicity. The advantages of such tests are reflected in the relatively affordable cost of cell cultures required for testing and measurement speed. However, the main limitation of *in vitro* research is the use of isolated cells, which excludes interaction with neighboring cells and intercellular signaling, which is necessary for the homeostasis of tissues and organs [12]. *In vivo* methods are applied to living organisms (mice, rats, and hamsters) and give the most reliable results in studying particle toxicity. The two main approaches are intratracheal inhalation and intratracheal instillation [13]. Applying the inhalation method requires the possession of expensive chambers in which the animals are exposed to certain particles. In contrast, the instillation method requires directly introducing particles into the trachea. Since the mentioned tests are performed on living organisms, they enable the most relevant results [14].

The OP cell-free assays are diverse and include dithiothreitol (DTT) assay, ascorbic acid (AA) assay, BPEA nitroxide assay, glutathione (GSH), dichlorofluorescein (DCFH) assays, p-hydroxyphenylacetic acid (POHPAA), and electron spin resonance (ESR). The DTT, GSH, and AA assays measure the depletion rate of chemical proxies for cellular reductants (DTT) or antioxidants (AA), which is proportional to the generation rate of ROS, while the ESR measures the generation of ROS via electron spin resonance. On the contrary, particle-bound ROS measurements, such as the DCFH assay and BPEAnit, use fluorescent-based techniques to measure concentrations of specific ROS, usually the hydroxyl radical or hydrogen peroxide in PM samples. These methods usually reveal a different response to the chemical constituents of PM, and they differ greatly in sensitivity and application. Further, environmental OP values can be expressed in two ways: as the OP normalized to the volume of air containing a certain concentration of PM, or as the OP normalized to the mass of particles,

representing the intrinsic OP of the PM analyzed. In addition to the variety of existing methods, it is also important to emphasize that there is no unique way of PM collection. The PM can be collected on different types of filters using high and low-volume samplers. This method's main disadvantages are poor particle recovery and particle aging, which leads to a certain kind of bias. In order to minimize sampling errors and obtain the most accurate experimental data, various methods of particle collection have been introduced, such as versatile aerosol concentration enrichment system (VACES) [15], particle into liquid sampler (PILS) [16], particle into liquid quencher (PINQ) [17], samplers based on the condensation of particles, and micro-orifice uniform deposition impactor (MOUDI) for size-segregated samples [18]. However, automation and online measurements have been a growing area of study, as they have the potential to significantly reduce analysis time, materials and would also help to obtain data with better time resolution [19-21]. Taking into account all aforementioned, these methods usually reveal a different response to the chemical constituents of PM, and they differ greatly in sensitivity and application. Also, chemical composition and physical properties could affect OP size distribution. Therefore, no method can define the total OP of measured PM, so it is always better to combine them.

### **What we know about source apportionment**

The major objective of SA models in the atmospheric sciences is to determine which specific sources of pollution contribute what percentage of total air pollution. In the literature, three different approaches to SA have been documented.

According to the first approach, the sources are identified, using different methods based on processing monitoring data. The way the data is processed with this approach generally results in minor mathematical artifacts, considering the simplicity of the data processing. With this method, for example, the location of specific sources of pollution can be determined by simply correlating wind speed with measured components [22]. The second approach is relatively more complex and enables simulating aerosol emission, generation, transport, and deposition using emission inventories and/or dispersion models [23]. Since an emission inventory is defined as the quantity of pollutants released into the atmosphere, which is not always available, a lack of this data limits these methods. Nevertheless, these methods may assist in identifying important sources of pollution, which may be significant for adopting various regulatory measures. Finally, the third approach represents statistical analyses of PM chemical data obtained from receptor locations (receptor models), and represents the most complex approach. Receptor modeling assumes mass and species conservation and uses a mass balance analysis to identify and apportion airborne PM sources. Therefore, the primary objective of receptor models is to identify the possible sources of PM and to obtain data on their contributions to the bulk PM mass. Even human exposure to these pollution components has been evaluated to assess their health effects and risks [24-26]. Thus the information provided by receptor models is key to the design of effective mitigation strategies on the local and meso-scale. In the literature, a variety of statistical models and modeling approaches depending on the knowledge required before receptor modeling can be found [27].

An important step in obtaining information that is crucial for developing air quality management strategies is the quantitative distribution of pollutants to their specific sources. Previous SA studies are based mostly on PM<sub>2.5</sub> and PM<sub>10</sub>, while the positive matrix factorization (PMF) and related methods, such as principle component analysis (PCA), multilinear regression (MLR), UNIMAX, and chemical mass balance (CMB), were the most frequently used. A large number of studies have established distributions for a restricted number of

chemical components included in particles, such as heavy metals, carbonaceous, some specific organic tracers and water-soluble species, in addition to PM distributions. Hopke *et al.* [28] summarized in SA review that research conducted on PM<sub>2.5</sub> and PM<sub>10</sub> samples had provided information on the apportionments that define nine pollution sources. These sources are defined as:

- *Inorganic source*: sulfate, nitrate, and mixed secondary inorganic aerosol (SIA).
- *Dust*: it is the collective term for crustal material, which includes both natural soil and desert dust.
- *Sea salt*: this source usually contains Na and Cl.
- *Traffic*: both exhaust and non-exhaust emissions.
- *Industry*: this source mainly contains different metals characteristic of areas burdened by different industrial activities.
- *Biomass burning*: mix of several different sources (cooking, heating, biomass burning...).
- *Coal/oil combustion*.
- *Other*: the SOA or uncalculated mass depending on the examined location/s.

Based on previous researches, the application of SA is important for several reasons. First of all, the results of SA allow insight into the dominant sources of pollution, which can help develop strategies and regulations that would contribute to improving air quality. Then, defining the source of pollution and knowing the concentrations of specific components characteristic of that source can help in health-oriented research.

### Source apportionment of oxidative potential

In contrast to many SA of PM studies conducted worldwide, there is a limited number of SA studies applied to OP. According to current literature data, about thirty studies dealt with this issue, tab. 1. Although this represents a small number of scientific papers that would allow writing a literature review, this paper aims to point out the importance of this topic and state the advantages and disadvantages of this approach.

The SA of estimated OP values resulting from field campaigns has shown differences among assays and sites studied, along with high temporal and spatial variability in each site. Studies conducted to date have examined the toxicity of PM collected mostly in urban areas, including locations impacted by nearby roads, airports, harbours, power plants, and specific local and regional sources. In contrast, few studies were conducted in rural and industrial sites [29-33]. Furthermore, the greatest emphasis was applied to PM<sub>2.5</sub>, while only a few studies investigated the association of OP of quasi-ultra-fine and coarse particles with potential sources [32, 34-41]. In support of the fact that these studies are still in the development phase and require greater attention, the fact that so far, the largest number of studies has been conducted in China, somewhat fewer in the USA, and only a few studies in Europe.

**Table 1. The SA of PM studies related to OP conducted worldwide**

	Type of site	PM	Sampling duration	Source contribution to OP
[42]	Peri-urban background and urban	2.5	Mar.-May 2012	Secondary aerosols, Zn source, Al source, Pb source, and Fe source
[29]	3 urban and 2 rural sites	2.5	June 2012-Sept. 2013	Biomass burning, secondary aerosols, vehicle, and mineral dust
[43]	Urban	2.5	June 2012-Apr. 2013	Biomass burning, light duty, and heavy duty vehicles

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**Table 1. Continuation**

[30]	7 different urban and rural	2.5	June 2012-Mar. 2013	Biomass burning, secondary aerosols, and traffic
[44]	3 urban	2.5	May 2015-Apr. 2016	Biomass burning, secondary aerosols, vehicles, coal combustion, and industry
[45]	Urban	2.5	Mar. 2012-Mar. 2013	Biomass burning, traffic, coal combustion, and waste incineration
[31]	Industrial	2.5	Jan.-Mar. 2018	Biomass burning, vehicles, crustal material, industry, secondary sulphate, and sea spray
[46]	Urban background	2.5	June-Sept. 2017 Feb.-Mar.2018	Biomass burning, secondary aerosols, and vehicles
[47]	Urban	2.5	May 2015-Apr. 2016	Traffic, dust, coal combustion, industry, and secondary sulphate
[48]	Urban	2.5	Sept. 2011-Aug. 2012	Biomass burning, secondary aerosols, secondary sulphate, and marine vessels
[49]	Urban	2.5	Apr.-Dec. 2017 May- June 2017	Vehicle, dust, coal combustion, industry, secondary sulphate, secondary nitrate, and metals
[50]	Suburban	2.5	winter and spring 2010-2011	Biomass burning, traffic, light and heavy duty vehicles, and industry,
[51]	Suburban	2.5	Mar.-Dec. 2016	Metals, aromatic substances, humic-like, and protein-like substances
[52]	Urban	2.5	Nov.-Dec. 2016	Biomass burning, secondary aerosols, vehicle, and land fossil fuel combustion
[53]	Urban	2.5	Sept. 2011-Avg. 2012	Biomass burning, secondary aerosols, vehicle, secondary sulphate, marine vessels, and metals
[54]	2 urban	2.5	15 days in Feb. 2019	Secondary aerosols, vehicle, and mixed sources
[55]	Rural	2.5	Aug. 2016 Jan.-Feb. 2017	Biomass burning, vehicle, mineral dust, coal combustion, secondary sulphate, and secondary nitrate
[33]	3 urban background, rural, and road	2.5	May 2018-May 2019	Biomass burning, secondary aerosols, vehicle, dust, secondary sulphate, ship emission, secondary nitrate, and agricultural emission
[56]	urban	2.5	Jan. 2 <sup>nd</sup> to 16 <sup>th</sup> Apr 7 <sup>th</sup> to 23 <sup>rd</sup> July 3 <sup>rd</sup> to 18 <sup>th</sup> Oct 12 <sup>th</sup> to 28 <sup>th</sup> , 2017	Primary biomass burning WSOC, other WSOC primary combustion, transition metal ions, biomass burning SOC, aromatic SOC, and the aged biogenic SOC
	Type of site	PM	Sampling duration	Source contribution to OP
[36]	2 urban and one urban background	<0.25	June-Oct. 2014	Secondary aerosols, light duty, and heavy duty vehicles
[34]	Urban-traffic and urban background	0.49	Jan.-Mar. 2013 May-July 2013	Biomass burning and traffic
[35]	Urban and suburban	0.18, 2.5	July 2012-Feb. 2013	Secondary aerosols, vehicles, wood smoke, crustal materials, and primary biogenic source
[38]	2 urban	1, 2.5	Nov. 2014, Jan.-Feb. 2015	Biomass burning, secondary aerosols, traffic, coal combustion, and cooking

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**Table 1. Continuation**

[37]	Urban	2.5, 2.5-10	July 2016 to July 2017	Biomass burning, low volatility oxygenated organic aerosol, and hydrocarbon-like organic aerosol
[32]	Urban, rural, road side, urban background	2.5, 10	Entire year 2013 Jan.-Sept. 2014	Biomass burning, secondary aerosols, traffic, and crustal material
[41]	Urban background	2.5, 10	Nov. 2016-Nov. 2017	Biomass burning, vehicle, crustal material, secondary sulphate, sea spray, secondary nitrate, and carbonates
[57]	Urban	10	Nov. 2013-Oct. 2014	Biomass burning and traffic
[39]	14 urban	10	2003-2018	Biomass burning, secondary aerosols, vehicles, dust, primary biogenic, and secondary nitrate
[40]	Urban	10	Dec. 2019-Mar. 2020 May-Aug. 2020	Secondary aerosols, traffic, soil and resuspended dust, and industry
[58]	Urban	TSP	Jan.-Mar., Apr.-June, Oct., Nov. 2016, Jan. 2017	Biomass burning, secondary sulphate, ship emission, and land fossil fuel combustion

In the majority of SA of OP studies, the OP was measured by applying the DTT assay. However, comparing the results obtained in different studies using this assay may be questionable, considering that there is no standardized protocol for the DTT assay. Filter extraction was mainly done by 30 minutes sonication in ultra-pure water, but the concentrations of DTT and DTNB were in the wide range. The DTT concentrations ranged between 0.1 mM to 10 mM, while the concentrations of DTNB were between 0.2 mM and 10 mM. Also, in most of the studies, before adding DTNB, trichloroacetic acid and Tris-HCl buffer containing EDTA were added, while in a few studies, this step was avoided. From the aforementioned, it can be concluded that one of the first steps is determining the detailed procedure for applying the DTT assay.

A detailed analysis of the existing literature showed that the largest contribution of ROS comes primarily from biomass burning, secondary organic aerosol and traffic/vehicles. Given that not all studies presented the contribution of a certain source of ROS in percentages, it can roughly be said that the association of biomass burning ranged from 9% to 97%. The SA of SOA's contribution was 6% to 67%, while the traffic/vehicles source fell between 16% to 88%. The greatest contribution of biomass burning to the overall OP was recorded during winter when the expected pollution was the highest. On the other hand, SOA and traffic were the most important contributing sources recorded during the summer months. Other sources that were also found to contribute to the OP were secondary sulfates, secondary nitrates and dust. Their contribution to OP was lower or equal to 30% collectively. Also, it was observed that depending on the characteristics of the measurement site and the influence of local sources, other potential sources contributing to higher concentrations of ROS were also determined (industry, ship emission, coal/oil combustion, crustal materials and, *etc.*). Daellenbach *et al.* [32] found that the major sources contributing to the mass and OP were different. The PM mass was mainly supplied by the secondary inorganic components, crustal material and secondary biogenic organic aerosols, while OP was mainly assigned to the fine-mode SOA from residential biomass burning and coarse-mode metals from vehicular non-exhaust emissions. At the urban roadside, OP PM<sub>10</sub> was found to be dominated by non-exhaust vehicular emissions (61%-88%), mainly in the coarse mode. At the rural background site, anthropogenic SOA dominated OP PM<sub>10</sub> (33%-67%). In more pristine environments, such as alpine valleys, which are strongly affected by residential heating during winter, bio-

mass burning organic aerosol (BBOA) was significant OP source in PM<sub>10</sub> fraction (7%-25%; 77%-97% during pollution events). A study conducted in France analysed PM<sub>10</sub> filters collected over 15 years at 14 different locations, including urban, traffic and Alpine valley site [39]. This study also highlights one aspect of SA OP complexity since the OP was measured using two different probes. Hence different types of chemical compounds were picked up with different probes and thus, different outcome from SA were established for the same samples. The primary road traffic, biomass burning, dust, methanesulfonic acid rich (MSA-rich), and primary biogenic sources had distinct positive redox activity towards the OP<sub>DTT</sub> assay, whereas biomass burning and road traffic sources only displayed significant activity for the OP<sub>AA</sub> assay. The daily median source contribution to the total OP<sub>DTT</sub> highlighted the dominant influence of the primary road traffic source. On the other hand, biomass burning and road traffic sources contributed evenly to the observed OP<sub>AA</sub>. A study conducted in an industrial area in Italy further confirmed that the SA of PM mass and OP do not necessarily overlap. For example, industry emission did not significantly contribute to the mass but had a significant contribution to the OP, whereas an opposite trend was observed for secondary sulphates [31]. Contrary to these findings, PMF results obtained at a rural site in Northwest China reported that biomass burning was among the major contributors to the mass and the OP in winter [55]. Secondary nitrates were a significant OP source during winter, while secondary sulphates dominated during summer. Altuwayjiri *et al.* [40] found that dust, SOA, local industrial activities, petroleum refineries, and traffic emissions were the four most significant identified sources of OP 31%, 20%, 19%, and 17%, respectively, in the arid area of Riyadh. Another study in Italy further confirmed that the SOA varies seasonally and that the SA of OP is different in different mass fractions, namely PM<sub>2.5</sub> and PM<sub>10</sub> [41].

The SA models also showed significant divergence in the contribution of different sources of OP vs. PM mass. In other words, OP and PM mass can have common contribution sources, but their contribution differs significantly. For example, Wang *et al.* [33] found that the contribution of agricultural activities at the rural site for PM<sub>2.5</sub> mass was 12% but for cellular OP was 62% at the same location. For this reason, the application of SA PM mass and OP does not necessarily lead to the same results, which is another indicator of the observation that the particular sources that contribute to a lesser extent to PM mass can overshadow the impact of that that source to their toxicity. It is necessary to examine the specific species that contribute to the increased OP to get a better insight into the specifics of SA of OP.

## Conclusion

Since PM toxicity is a poorly understood phenomenon, the first step in the holistic understanding of PM toxicity could be measuring OP of PM and their connection with specific sources. Detailed analyses, with a well-designed and uniform strategy of sampling, measurement, and application of a certain SA technique, could significantly help in understanding this issue. However, regardless of the advantages of this approach, certain limitations still need to be overcome to improve future research. Primarily it refers to standardizing sample extraction and acellular assays to determine OP. Also, when filters are analyzed, it is known that particles, that may carry short-lived ROS, are mostly gone from the sample. In light of this, it appears that a combination of online and offline methods is best suitable for understanding the various ROS. One should then, consider the uncertainties arising from the application of the SA model. Even though it is known that PM<sub>2.5</sub> penetrates to the deeper parts of the lungs and has a greater impact on human health, it is necessary to examine the effects of PM<sub>10</sub> in more detail.



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