Transient plasma-enhanced remediation of nanoscale particulate matter in restaurant smoke emissions via electrostatic precipitation

Sisi Yang[a], Patrick Ford[d], Sriram Subramanian[c], Dan Singleton[d], Jason Sanders[d], Stephen B. Cronin[a,b,d,*]

[a] Department of Physics and Astronomy, University of Southern California, Los Angeles, CA 90089, USA
[b] Ming Hsieh Department of Electrical Engineering, University of Southern California, Los Angeles, CA 90089, USA
[c] Daniel J. Epstein Department of Industrial & System Engineering, University of Southern California, Los Angeles, CA 90089, USA
[d] Transient Plasma Systems, Inc., Torrance, CA 90501, USA

ABSTRACT

It is now recognized that nanoscale particulate matter (PM) represents a substantial health hazard for our society, including PM from restaurant smoke. In this study, we explored the use of a transient pulsed plasma in conjunction with an applied DC bias to treat oil aerosols that closely resemble restaurant (i.e., charbroiler) smoke emissions. For polyaromatic olefin PAO-4 and soybean oil, we found that a three-order-of-magnitude reduction in particulates (i.e., 99.9% remediation) could be achieved with this system. Here, the plasma discharge was produced in a 4-in.-diameter cylindrical reactor with a 5–10 ns high voltage (30 kV) pulse generator together with applied DC bias voltages up to 10 kV. The distribution of nanoparticle sizes was measured using a scanning mobility particle sizer (SMPS) with diameter centered around 225 nm. Here, the main mechanism of remediation occurs in a two-step process in which the oil nanoparticles are first ionized by the free electrons and free radicals in the plasma and then the charged particles are swept out to the sidewalls of the reactor by the applied DC potential. We believe this general approach opens up new degrees of freedom in the design of electrostatic oil aerosol pollution control devices.

© 2020 Chinese Society of Particuology and Institute of Process Engineering, Chinese Academy of Sciences. Published by Elsevier B.V. All rights reserved.

Introduction

The wide acceptance of the serious health effects associated with nanoscale particulate matter (PM) produced by fossil fuel combustion has led to a wide range of nanotoxicology studies of the environmental emissions from restaurants in commercial cooking processes (e.g., charbroiling) (Chow et al., 2006; Dockery et al., 1993; Kaltsonoudis et al., 2017; McDonald et al., 2003; Oberdörster, Oberdörster, & Oberdörster, 2005; Pope et al., 2002; Samet, Dominici, Curriero, Coursac, & Zeger, 2000; Yancey, Apple, & Wharton, 2016). In 1997, the South Coast Air Quality Management District (SC-AQMD) in Southern California passed RULE 1138, which regulates PM emissions from chain-driven (i.e., conveyor-belt) charbroilers (Rule 1138, 1997). This PM is made up of oil aerosol particles approximately 100–200 nm in size, which are produced from fat-containing meat during the cooking process. In response to this ruling, these chain-driven charbroilers are now outfitted with high temperature oxidation catalysts located just above the hot cooking surface, mitigating these nanometer-scale oil aerosol particles. While chain-driven charbroilers are primarily used in large fast-food restaurants, a majority (~85%) of total restaurant smoke emissions are produced by open underfire charbroilers (Perryman, 2009; Whynot, Quinn, Perryman, & Votlucka, 1999). In a 2016 report, there was an estimated 1400 tons of particulate matter produced annually in New York city originating from these open-underfire charbroilers. According to the Department of Health and Mental Hygiene, it is estimated that more than 12% of premature deaths due to $PM_{2.5}$ (i.e., particles ≤2.5 μm) are attributed to open underfire charbroiler emissions (DEP, 2016; New York City Department of Environmental Protection, 2016). By equipping all of the restaurant charbroilers in New York city with effective pollution control technologies, an estimated 88% of these premature deaths can be reduced by limiting the $PM_{2.5}$ concentrations in the region (DEP, 2016).

* Corresponding author at: Ming Hsieh Department of Electrical Engineering, Department of Physics and Astronomy, Department of Chemistry, University of Southern California, Los Angeles, CA 90089, USA.
E-mail address: scronin@usc.edu (S.B. Cronin).

With the exception of New York city, San Francisco and San Joaquin, particulate matter (PM) emissions from open underfire charbroilers are largely unregulated and account for 94% of all restaurant emissions in these regions (Wright, 2009). A typical charbroiler produces around 10 lbs/day of particulates at volumetric flow rates above 1600 ft³/min. Per hamburger, this corresponds to about 5 g of particulate matter (PM) (Bose, 2012). It is important to point out that the high temperature oxidation catalysts in chain-driven charbroilers cannot be used to treat open underfire charbroilers because, in this configuration, the ventilation hood is typically more than 1 m above the cooking surface. Here, the exhaust is relatively cool, and the catalyst would need to be heated separately in order to provide effective remediation. Currently available methods for removing oil aerosol particulates include electrostatic precipitation (ESP), wet scrubbers, and filtration (Gysel et al., 2018; Lee et al., 2011). For heavy grease cooking, such as hamburger charbroiling, filtration is cost-prohibitive and requires daily maintenance. In this approach, several filters are arranged in series, creating a large pressure drop making it necessary to use a high-power fan in order to achieve sufficiently high flow rates to reach kitchen ventilation compliance.

While there have been many reports characterizing harmful emissions from various cooking processes (Amouei Torkmahalleh, Gorjinezhad, Unluevcek, & Hopke, 2017; Buonanno, Johnson, Morawska, & Stabile, 2011; Gao et al., 2013; Kaltsonoudis et al., 2017; McDonald et al., 2003; Robinson et al., 2018; Schauer, Kleeman, Cass, & Simoneit, 2002; Yancey et al., 2016), there have been relatively few reports on techniques for remediating these harmful pollutants (Chang, Mi, Lin, Hsieh, & Chao, 2011; Gysel et al., 2018; Lee et al., 2011; Yang et al., 2019). Lee et al. reported emission rates and removal efficiencies of particulate matter by electrostatic precipitators (without plasma) in under-fired charbroilers with efficacies between 55%–97% (Lee et al., 2011). Researchers at the University of California, Riverside's commercial test cooking facility performed a comparative study of three remediation technologies using the South Coast Air Quality Management District (SCAQMD) Method 5.1. Of the three devices studied, a dual-stage filtration system, a device based on evaporative cooling and electrostatic ionization, and an electrostatic precipitator, only the electrostatic precipitator provided remediation above 80% (Gysel et al., 2018). Chang et al. reported plasma-based removal of gaseous polycyclic aromatic hydrocarbons from cooking fumes (Chang et al., 2011). In 2019, Yang et al. reported first results on plasma-based remediation of nanoscale particulate matter in restaurant smoke emissions, however, without an applied DC bias (Yang et al., 2019).

In the work reported here, we use a nanosecond pulse plasma in conjunction with a DC bias, which provides an electrostatic field across the reactor and produces a higher plasma density than the nanosecond pulsed plasma alone. When the DC bias voltage is added to the nanosecond pulse, the voltages sum additively, resulting substantially higher peaks fields. For example, a 30-kV nanosecond pulse applied with a 10-kV DC bias will provide a peak voltage of 40 kV. Here, the remediation occurs in a two-step process where the nanoparticles are ionized by free radicals in the plasma and then swept out to the sidewalls by the applied DC bias.

Experimental methods

In the study presented here, we use an oil aerosol generator from Aerosol Technologies International (ATI, Inc), which is created by forcing compressed air through a Laskin nozzle (Drew, Berntein, & Sidney, 1978; First, Rudnick, & Yan, 1992; Hinds, Macher, & First, 1983). Our plasma-based flow reactor consists of a 4-ft-long, 4-in.-diameter stainless steel cylindrical anode with a 25-mil single-wire cathode arranged in a coaxial geometry, as illustrated in Fig. 1. This system has electrical feedthroughs on either end of the reactor, one for supplying high DC voltages and the other for high voltage nanosecond pulses, as indicated in Fig. 1(a). Fig. 1(c) shows a circuit diagram illustrating how the DC bias is configured together with the nanosecond pulse generator. In this configuration, a high voltage capacitor protects the nanosecond pulse generator from the high voltage DC power supply, and an inductor protects the DC power supply from the nanosecond high voltage pulses. The
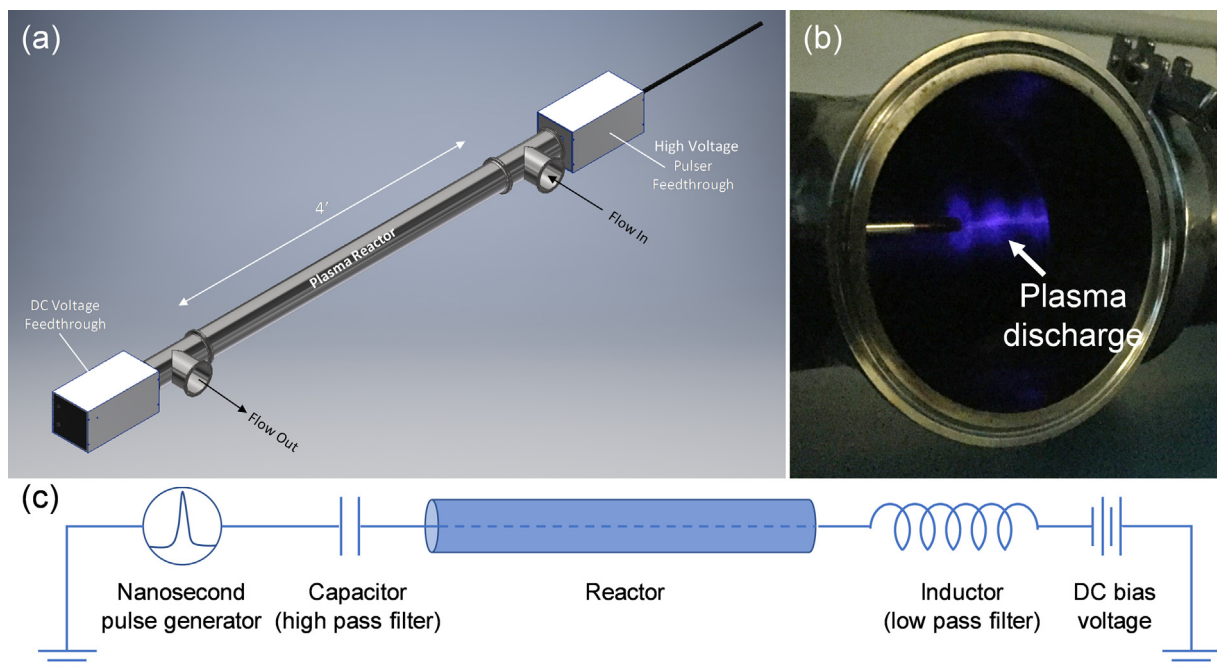

Fig. 1. (a) CAD drawing of the plasma-based reactor for restaurant particulate emissions remediation. (b) Photograph of the plasma discharge at the output port of the reactor system. (c) Schematic circuit diagram of the experimental setup.
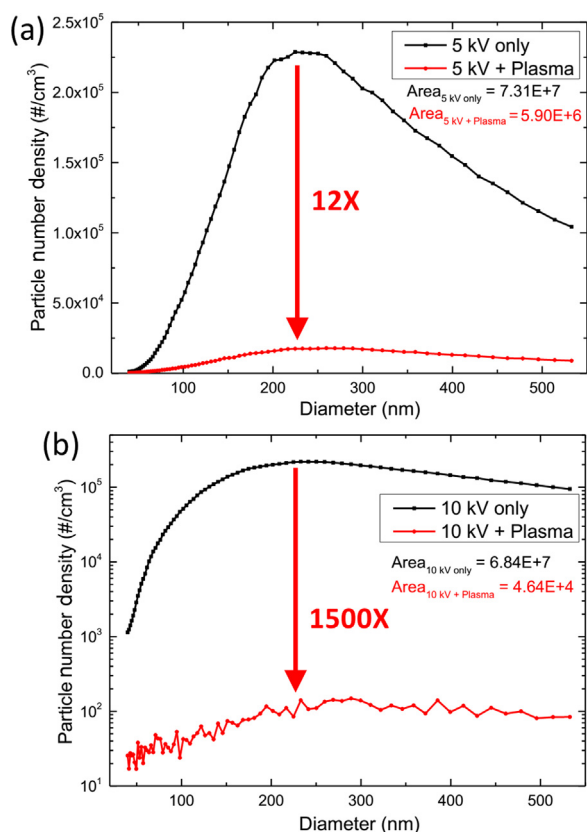
Fig. 2. Particle size distributions obtained with PAO-4 under applied DC voltages of (a) 5 kV and (b) 10 kV both with and without the 30 kV nanosecond pulsed plasma (pulse repetition rate of 200 Hz and electrically continuous power of 30 W).

Fig. 3. Particle size distributions obtained with soybean oil under applied DC voltages of (a) 2.5 kV and (b) 5 kV both with and without the nanosecond pulsed plasma running at a peak voltage of 30 kV, pulse repetition rate of 200 Hz, and continuous power of 30 W.

plasma is produced using a TPS Model 30X pulse generator operating at a peak voltage of 30 kV, a pulse repetition rate of 200 Hz, and a continuous power of 30 W. A typical waveform of the nanosecond high voltage pulse is plotted in Figure S1 in the Supplemental Information. Here, the generation of plasma is assisted by 10 kV DC power supply capable of supplying up to 30 W of continuous power. Particle size distributions (i.e., histograms) were measured using a scanning mobility particle sizer (SMPS) spectrometer (TSI Model 3938) with a condensation particle counter (CPC), capable of measuring particle distributions over the range from 14 to 685 nm. Baseline particle distributions (i.e., without plasma) exhibit highly stable distributions, as shown in Figure S2 in the Supplemental Information.

Results and discussion

Fig. 2(a) shows the particle size distributions taken under an applied DC voltage of 5 kV both with and without the nanosecond pulse generator running at a peak voltage of 30 kV, pulse repetition rate of 200 Hz, and continuous electrical power of 30 W. A comparison of these two distributions shows a 12-fold reduction of total PM concentration (i.e., 92% remediation). Here, the integrated areas are indicated in the plot corresponding to the total particle concentrations both with and without the transient pulsed plasma. Similarly, Fig. 2(b) shows the particle size distributions taken with an applied DC voltage of 10 kV both with and without a nanosecond pulse generator, exhibiting a 1500-fold reduction in PM concentration (i.e. 99.9% remediation). It should be noted that the particle distributions taken with 5 kV DC and 10 kV DC only, without the nanosecond pulse generator, are nearly identical to the untreated baseline data (i.e., no remediation) plotted in Figure S3 in the Sup-
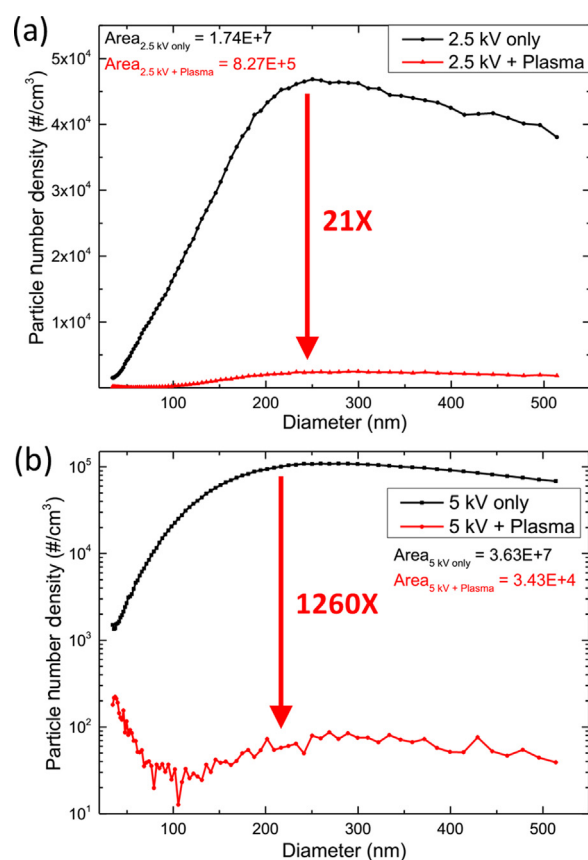
plemental Information. Here, we believe the main mechanism of remediation occurs in a two-step process in which the oil-based nanoparticles are first ionized by the high energy electrons and free radicals in the plasma and then the charged particles are swept out to the sidewalls of the reactor by the applied DC bias.

We can make a rough estimate for the time required for a 225-nm-diameter nanoparticle to be swept out to the sidewalls of the reactor by assuming an electrostatic force of qE accelerating a nanoparticle of mass $\rho \cdot \frac{4}{3}\pi\left(\frac{d}{2}\right)^3$, resulting the formula $t = \sqrt{\frac{\rho R \pi d^3}{3qE}}$, where d, ρ and q are the diameter, density, and charge of the nanoparticle, respectively, E is the electric field, and R is the radius of the reactor. For a 4-in.-diameter reactor under an applied DC bias of 10 kV, a 225-nm-diameter nanoparticle with a charge of one electron (i.e., minimum charge) and mass density of 0.819 g/mL (density of PAO-4), this "sweep out" time is 79 msec. The timing and kinetics of the soybean oil particles are expected to be quite similar to those of the PAO-4.

We have also performed a separate set of measurements using soybean oil rather than PAO-4. The soybean oil more closely resembles the oil-based nanoparticles that are generated by the charbroiling of hamburger meat and is often used as a surrogate grease generator following the UL 1046 standard method (UL, 2010). However, it is also worth noting that these soybean oil grease aerosol particles are generated at room temperature. Fig. 3 shows particle size distributions taken with applied DC voltages of 2.5 kV and 5 kV both with and without the nanosecond pulsed plasma. For a DC bias of 2.5 kV, we observed a 21-fold reduction in PM concentration (i.e., 96% remediation). For a DC bias of 5 kV DC, we observe

a 1260-fold reduction in PM concentration (i.e., 99.9% remediation). The improved remediation obtained with a DC bias of 5 kV compared to that of 2.5 kV DC can be attributed to the increased electric fields that are achieved when adding the 30 kV peak pulse voltage. Also, at higher DC biases, the plasma density is higher and fills a more substantial volume of the reactor. It should be noted that the distributions observed with 2.5 and 5 kV DC bias only (i.e., without the nanosecond pulse generator) are nearly identical to the untreated data (i.e., no remediation). As was the case for PAO-4, the main mechanism of remediation, here, occurs in a two-step process in which the oil nanoparticles are first ionized by the free electrons and free radicals in the plasma, and then the charged particles are swept out to the sidewalls of the reactor by the applied DC potential.

It should be noted that the transient nature of the plasma dictates that very little current is drawn in creating the plasma. That is, once the plasma is created, the applied electric field collapses before a substantial amount of current (and hence electric power) can flow. Because of its transient nature, this is a non-thermal cold plasma with electron energies around 30 eV ($T=10^5$ K) and vibrational modes of the molecules remaining near room temperature. These "hot" electrons are responsible for providing new chemical pathways by forming charge-free radicals and highly reactive species in the plasma, which include atomic oxygen and ozone, driving chemical reactions in a fundamentally different manor than standard equilibrium chemistry. Previously, Yang et al. showed that oil aerosol particles produced during the charbroiling process were shifted to smaller diameters when treated with transient pulsed plasma (without DC bias), likely due to chemically active free radicals, such as atomic oxygen, which break down the grease particles into CO, CO_2 and other hydrocarbons similar to the plasma-induced break down of polymer films (Yang et al., 2019). Future studies are needed in order to obtain a complete understanding of the chemical pathways associated with this aspect of the remediation process. As mentioned above, our plasma-based flow reactor consists of a 4-ft-long, 4-in.-diameter stainless steel cylindrical. Assuming a pressure drop of 1–2 in. of water, this reactor will operate up to volumetric flow rates of 850–1250 CFM. Typical commercial kitchen ventilation systems start around 2000–3000 CFM, thus requiring 2–4 of these reactors in parallel.

Conclusion

In conclusion, we demonstrate the effectiveness of a transient pulsed plasma to improve the remediation efficiency of oil-based nanoparticles through electrostatic precipitation by several orders of magnitude (99.9% remediation). Here, a non-thermal (i.e., cold) plasma is generated using 30-kV pulses with 5–10 nsec rise times. The high energy electrons and free radicals in this plasma ionize these oil-based particulates, which are subsequently swept out to the sidewalls of the reactor under an applied DC electric field on msec timescales. In addition to providing an electrostatic field across the reactor, the applied DC voltage sums additively with the nanosecond high voltage pulse creating an ionizing plasma over a larger volume of the reactor than that created with the nanosecond high voltage pulse alone. In this proof-of-principle demonstration, oil-based nanoparticles are generated using a Laskin nozzle aerosol generator using polyaromatic olefin PAO-4 and soybean oil, which is a common surrogate for restaurant smoke emissions. This combined approach presents new design considerations in the treatment and optimization of oil-based aerosol particulates produced by restaurant smoke emissions, which present a substantial health hazard to our society.

Declaration of interests

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.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.partic.2020.06.003.

References

Amouei Torkmahalleh, M., Gorjinezhad, S., Unluevcek, H. S., & Hopke, P. K. (2017). Review of factors impacting emission/concentration of cooking generated particulate matter. *Science of the Total Environment, 586*, 1046–1056.

Bose, L. (2012). Controlling the emissions of charbroiled burgers. In *UCR today*. Riverside: University of California. UCR Today

Buonanno, G., Johnson, G., Morawska, L., & Stabile, L. (2011). Volatility characterization of cooking-generated aerosol particles. *Aerosol Science and Technology, 45*, 1069–1077.

Chang, H. C., Mi, H. H., Lin, Y. C., Hsieh, L. T., & Chao, H. R. (2011). Removal of gaseous polycyclic aromatic hydrocarbons from cooking fumes using an atmospheric plasma reactor. *Journal of Environmental Science and Health, Part A, 46*, 1443–1449.

Chow, J. C., Watson, J. G., Mauderly, J. L., Costa, D. L., Wyzga, R. E., Vedal, S., et al. (2006). Health effects of fine particulate air pollution: Lines that connect. *Journal of the Air & Waste Management Association, 56*, 1368–1380.

DEP. (2016). *Emissions control devices for commercial char broilers.* Department of Environmental Protection: NYC Rules.

Dockery, D., Pope, C., Xu, X., Spengler, J., Ware, J., Fay, M., et al. (1993). An association between air pollution and mortality in six U.S. Cities. *The New England Journal of Medicine, 329*, 1753–1759.

Drew, R. T., Berntein, D. M., & Sidney, L. (1978). The Laskin aerosol generator. *Journal of Toxicology and Environmental Health, 4*, 10.

First, M. W., Rudnick, S. N., & Yan, X. (1992). Use of alternative liquids in Laskin nozzle generated aerosols for filter testing. *American Industrial Hygiene Association Journal, 53*, 6.

Gao, J., Cao, C., Wang, L., Song, T., Zhou, X., Yang, J., et al. (2013). Determination of size-dependent source emission rate of cooking-generated aerosol particles at the oil-heating stage in an experimental kitchen. *Aerosol and Air Quality Research, 13*, 488–496.

Gysel, N., Welch, W. A., Chen, C.-L., Dixit, P., Cocker, D. R., & Karavalakis, G. (2018). Particulate matter emissions and gaseous air toxic pollutants from commercial meat cooking operations. *Journal of Environmental Sciences, 65*, 162–170.

Hinds, W. C., Macher, J. M., & First, M. W. (1983). Size distribution of aerosols produces by the Laskin aerosol generator using substitute materials for DOP. *American Industrial Hygiene Association Journal, 44*, 6.

Kaltsonoudis, C., Kostenidou, E., Louvaris, E., Psichoudaki, M., Tsiligiannis, E., Florou, K., et al. (2017). Characterization of fresh and aged organic aerosol emissions from meat charbroiling. *Atmospheric Chemistry and Physics, 17*, 7143–7155.

Lee, J.-B., Kim, K.-H., Kim, H.-J., Cho, S.-J., Jung, K., & Kim, S.-D. (2011). Emission rate of particulate matter and its removal efficiency by precipitators in under-fired charbroiling restaurants. *The Scientific World Journal, 11*, 1077–1088.

McDonald, J. D., Zielinska, B., Fujita, E. M., Sagebiel, J. C., Chow, J. C., & Watson, J. G. (2003). Emissions from charbroiling and grilling of chicken and beef. *Journal of the Air & Waste Management Association, 53*, 185–194.

(2016). *New York City administrative code sections 24-105 and 24-149.4.* New York City Department of Environmental Protection.

Oberdörster, G., Oberdörster, E., & Oberdörster, J. (2005). Nanotoxicology: An emerging discipline evolving from studies of ultrafine particles. *Environmental Health Perspectives, 113*, 823–839.

Perryman, P. (2009). *Control of emissions from restaurant operations.* South Coast Air Quality Management District.

Pope, C. A., III, Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., et al. (2002). Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *JAMA: The Journal of the American Medical Association, 287*, 1132–1141.

Robinson, E. S., Gu, P., Ye, Q., Li, H. Z., Shah, R. U., Apte, J. S., et al. (2018). Restaurant impacts on outdoor air quality: Elevated organic aerosol mass from restaurant cooking with neighborhood-scale plume extents. *Environmental Science & Technology, 52*, 9285–9294.

Rule 1138. (1997). *Control of emissions from restaurant operations.* South Coast Air Quality Management District.

Samet, J. M., Dominici, F., Curriero, F. C., Coursac, I., & Zeger, S. L. (2000). Fine particulate air pollution and mortality in 20 U.S. cities, 1987–1994. *The New England Journal of Medicine, 343*, 1742–1749.

Schauer, J. J., Kleeman, M. J., Cass, G. R., & Simoneit, B. R. T. (2002). Measurement of emissions from air pollution sources. 4. C-1-C-27 organic compounds from cooking with seed oils. *Environmental Science & Technology, 36*, 567–575.

UL. (2010). *1046 standard for grease filters for exhaust ducts*.

Whynot, J., Quinn, G., Perryman, P., & Votlucka, P. (1999). Control of fine particulate (PM2.5) emissions from restaurant operations. *Journal of the Air & Waste Management Association, 49*, 95–99.

Wright, S. R. (2009). *Retrofit air pollution control filter for restaurant underfired charbroilers*. U. S. Environmental Protection Agency.

Yancey, J. W. S., Apple, J. K., & Wharton, M. D. (2016). Cookery method and endpoint temperature can affect the warner-bratzler shear force, cooking loss, and internal cooked color of beef semimembranosus and infraspinatus steaks. *Journal of Animal Science, 94*, 4434.

Yang, S., Subramanian, S., Singleton, D., Schroeder, C., Schroeder, W., Gundersen, M. A., et al. (2019). First results on transient plasma-based remediation of nanoscale particulate matter in restaurant smoke emissions. *Environmental Research, 178*.