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Characterization of Primary Organic Aerosol Emissions from Meat Cooking, Trash Burning, and Motor Vehicles with High-Resolution Aerosol Mass Spectrometry and Comparison with Ambient and Chamber Observations

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Organic aerosol (OA) emissions from motor vehicles, meat-cooking and trash burning are analyzed here using a high-resolution aerosol mass spectrometer (AMS). High resolution data show that aerosols emitted by combustion engines and plastic burning are dominated by hydrocarbon-like organic compounds. Meat cooking and especially paper burning emissions contain significant fractions of oxygenated organic compounds; however, their unit-resolution mass spectral signatures are very similar to those from ambient hydrocarbon-like OA, and very different from the mass spectra of ambient secondary or oxygenated OA (OOA). Thus, primary OA from these sources is unlikely to be a significant direct source of ambient OOA. There are significant differences in high-resolution tracer *m/z*s that may be useful for differentiating some of these sources. Unlike in most ambient spectra, all of these sources have low total *m/z* 44 and this signal is not dominated by the CO_2^+ ion. All sources have high *m/z* 57, which is low during high OOA ambient periods. Spectra from paper burning are similar to some types of biomass burning OA, with elevated *m/z* 60. Meat cooking aerosols also have slightly elevated *m/z* 60, whereas motor vehicle emissions have very low signal at this *m/z*.

Introduction

Organic aerosols (OA) constitute a significant fraction of ambient particulate matter (PM) (*1*) and have important effects on human health and climate. The sources of OA remain uncertain. Source apportionment techniques use chemical composition measurements to quantify the fraction of ambient OA originating from different sources. The chemical mass balance of organic molecular markers (CMB-OMM) has dominated OA source apportionment for more than a decade (*2, 3*). The large amount of mass required and high cost of analysis lead to poor temporal resolution (hours to days). Chemical analyses with gas chromatography–mass spectrometry (GC–MS) can only identify about 10% of the organic mass in ambient particles (*4*). Real-time aerosol mass spectrometers produce OA mass spectra with high time resolution. The Aerodyne aerosol mass spectrometer (AMS) is the most widely used instrument of this type and can produce quantitative data for OA concentrations (*5*). Zhang et al. (*6, 7*) showed that statistical component analysis of AMS spectra could be used to quantify the contribution of hydrocarbon-like (reduced) OA (HOA) and oxygenated OA (OOA) in Pittsburgh. HOA was interpreted as the sum of primary OA (POA) sources, whereas OOA was interpreted as SOA. Recently several other statistical methods have been applied to AMS spectra, confirming and extending the Pittsburgh results to many locations while identifying more detail in the source apportionment (*1, 8, 9*). Interpretation of the component spectra from statistical analyses requires comparison with spectra obtained from different sources (*7*). A public web-based AMS mass spectral database (AMS-MSD) has been established for this purpose (*9*). Currently all spectra in this database are unit mass resolution (UMR), i.e., they only report total signal intensity at each integer *m/z*. The recently developed high-resolution time-of-flight AMS (HR-ToF-AMS) (*10*) separates ions of different elemental compositions at each *m/z* which should allow improved characterization of OA sources and component analysis with statistical techniques (*9*).

Mobile sources such as gasoline and diesel-powered vehicles contribute to ambient OA concentrations, especially in urban areas (*6*). A few UMR AMS spectra from these sources are available in the AMS-MSD. The acquisition of HR spectra from vehicles is of great interest for statistical analyses (*9*) using HR data.

Previous studies have reported that meat-cooking operations contribute significantly to urban OA concentrations, for example ref *11*. A back-of-the envelope estimate for the Los Angeles area (Supporting Information (SI) Table SI-1) indicates the potential importance of meat cooking as a source. More than 120 compounds are quantified in meat cooking OA, prominent among which are palmitic acid, stearic acid, oleic acid, and cholesterol. Systematic and significant inconsistencies between ambient molecular marker data and published source profiles highlight the need for additional research (*12*).

Trash burning can also be an important OA source, especially in developing countries. Currently, this source is not included in most emission inventories, and there are only a few studies available. Source apportionment for the ACE–Asia experiment suggests a refuse burning contribution ranging from ~1 to 24% of the extractable organics (*13*). Lemieux et al. (*14*) characterized and quantified a waste stream for New York State residents, where paper (61.7%) and plastic (7.6%) make up the biggest fractions. To our knowledge, AMS mass spectral signatures of meat cooking or trash burning have not been reported.

The purpose of this work is to characterize the composition and high-resolution electron-impact (EI) mass spectral signatures of OA emissions from meat cooking, trash burning, and combustion engines. Particles from several sources were

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analyzed with an HR-ToF-AMS after dilution. Mass spectral signatures and marker ions are compared with ambient and smog chamber data.

Experimental Section

The AMS instruments and data analysis techniques have been described elsewhere (5, 10). We used a HR-ToF-AMS, operated only in the "W" highest resolution mode (10) that allows the determination of the elemental composition of most ions.

The experiments were carried out in a large shed (volume: ~240 m^3) in the town of Niwot, 10 miles northeast of Boulder, Colorado, approximately one mile from a local highway. An inlet was placed near the ceiling of the shed through which air was drawn at 10 Lpm through 3/8 in. copper tubing and split into the different instruments (inlet residence time ~ 0.9 s). Sampling from this location resulted in both high and consistent dilution factors. The dilution factors from the raw exhausts were ~100−1000 for the data presented here, and where determined by comparing the CO, particle mass, and/ or CO$_2$ concentration directly at the source vs in the sampling system. The shed was not hermetically sealed, leading to dilution of particle concentrations with ambient air (which had very low PM concentrations ~1 μg m^{-3} OA) on a time scale of ~20 min. The experiments were carried out by producing high PM loadings (putting one piece of meat on the grill, burning one piece of trash, or revving up an engine) which were vigorously mixed using several large fans while sampling continuously for up to 30 min. Between experiments, PM concentrations were decreased to ambient levels by shed dilution or opening the shed doors prior to starting the next experiment. The experiments were carried out late April/early May 2007, with ambient temperature averaging 15 °C (range 4−30 °C).

Ten meat cooking, six engines, and four trash burning sources were tested in these experiments (see SI Tables SI-2−4). All meats were cooked on a Pacific Gas Specialties propane grill. Briquettes made of compressed pumice rocks, located between flame and grill, were used to distribute the heat. To evaluate the influence of different cooking methods, one type of meat (lean burgers) were also cooked in a pan over a propane flame, in order to prevent fat from dripping into the flames. After each experiment, the grill was moved out of the shed and cleaned. All "trash" materials were burned inside a clean 55 gallon metal drum. The vehicles and grass trimmer were warmed up outside the shed, either briefly operated inside the shed or operated outside the shed with their exhaust directed toward a gap in the door, and turned off after sufficient emissions were collected in the shed.

Results and Discussion

As examples of the typical experimental procedure, the time traces of measurements from various sources are shown in SI Figure SI-1. The particle concentrations typically reached a peak and then declined on a time scale of ~20 min mainly due to dilution. The contribution of inorganic species was far smaller (less than 1%) than the organic contribution in all cases. Depending on the source, the produced PM concentrations varied significantly. Emission ratios of PM to CO$_2$ were determined for all sources and are of the same order-of-magnitude of those reported in the literature for motor vehicle sources (e.g. ref (15)). They are included in SI Tables SI-2−4 and Figure SI-2 only to provide context for these experiments, and they should not be used to estimate emissions from these types of sources since the experimental setup was not designed for accurate quantification. There were large differences among different types of meat when cooked in the same manner, with increasing emissions as the fat content increased, which is qualitatively consistent

with findings from other studies (16−18). As described by Rogge et al. (19), cooking meat on a grill is a very efficient way to produce aerosols composed of fatty acids since liquid grease droplets fall into the gas flame or onto the heated briquettes where they can vaporize and quickly renucleate and grow into small particles.

Diesel vehicles in our study produced higher organic emissions than gasoline-powered vehicles, again consistent with previous studies (15). The burning of polystyrene (PS) visibly produced much larger amounts of black carbon (BC) than organics. BC is formed in flames by the growth of large polycyclic aromatics and we speculate that the presence of aromatic styrene monomers and chains may favor BC instead of OA formation in PS combustion.

Mass Spectral Signatures. UMR organic mass spectra (MS) were extracted from the AMS data (20). Selected MS from various sources as well as typical ambient HOA and OOA spectra (6) are presented in Figure 1 (for the complete set of MS, see SI Figure SI-3). For sources with very high emissions, spectra from peak concentrations and from periods when concentration values were 10−20 μg m^{-3} have very similar patterns (not shown), often with a slight shift toward higher m/zs for spectra at high concentrations for several of the meat sources. The spectra from the sources studied here are much more similar to ambient HOA than OOA spectra, and thus these sources do not directly emit "ambient OOA."

All the meat cooking spectra (Figure 1a−d) show a similar pattern, especially for $m/z < 100$. The most prominent spectral fragment is m/z 55. The meat cooking spectra are dominated by the following UMR ion series: $C_nH_{2n+1}^+$ and $C_mH_{2m+1}CO^+$ (m/z 29, 43, 57, 71, 85...), and $C_nH_{2n-1}^+$ and $C_mH_{2m-1}CO^+$ (m/z 41, 55, 69, 83...). These ion series typically result from EI of saturated alkanes and alkenes plus cycloalkanes, respectively, and also from oxygenated species such as organic acids. However, the abundance of fragments $m/z >$ 44 indicates that the molecules that constitute meat cooking OA have a large alkyl backbone with limited oxygen, consistent with the species identified by GC−MS (19, 21). Spectra from meats with highest fat content, including salmon, also have the highest fraction of the total signal at high m/z ratios, consistent with a high content of molecules with long alkyl-chains in fats.

MS from two motor vehicles (one gasoline- and one diesel-powered) are presented in Figure 1e−f. OA from diesel exhaust is typically dominated by recondensed engine lubricating oil and consists mainly of n-alkanes, branched alkanes, cycloalkanes, and aromatics (15). The motor vehicle spectra are dominated by the same ion series as the meat cooking spectra, but the patterns differ slightly: nearly all vehicles have the most prominent peak at m/z 57, characteristic of saturated hydrocarbons.

Figure 1g−h show the MS from plastic no. 2 and paper burning, respectively. Plastic nos. 2 and 4 (SI Figure SI-3) spectra have very similar patterns compared to each other, with the most prominent peaks being at m/z 57 and 55, respectively. Both are dominated by the same UMR ion series as spectra from meat cooking and the motor vehicles. Plastic no. 6 showed a very different MS pattern than the other plastics, with a dominant peak for styrene, but also with large signals beyond m/z 44. The UMR MS from paper burning is again similar to the MS from meat cooking and motor vehicles. Its most prominent peak is at m/z 57, followed by m/z 59 and it has significant signal up to m/z 150. However, it also contains large signals at m/z 60 and 73 which are also observed in the spectra of biomass burning aerosols (22). This is not surprising since cellulose is a sugar polymer that contains abundant oxygen, and since species like levoglucosan ($C_6H_{10}O_5$) that arise from the pyrolysis of cellulose give rise to intense signals at m/z 60 and 73 (22).
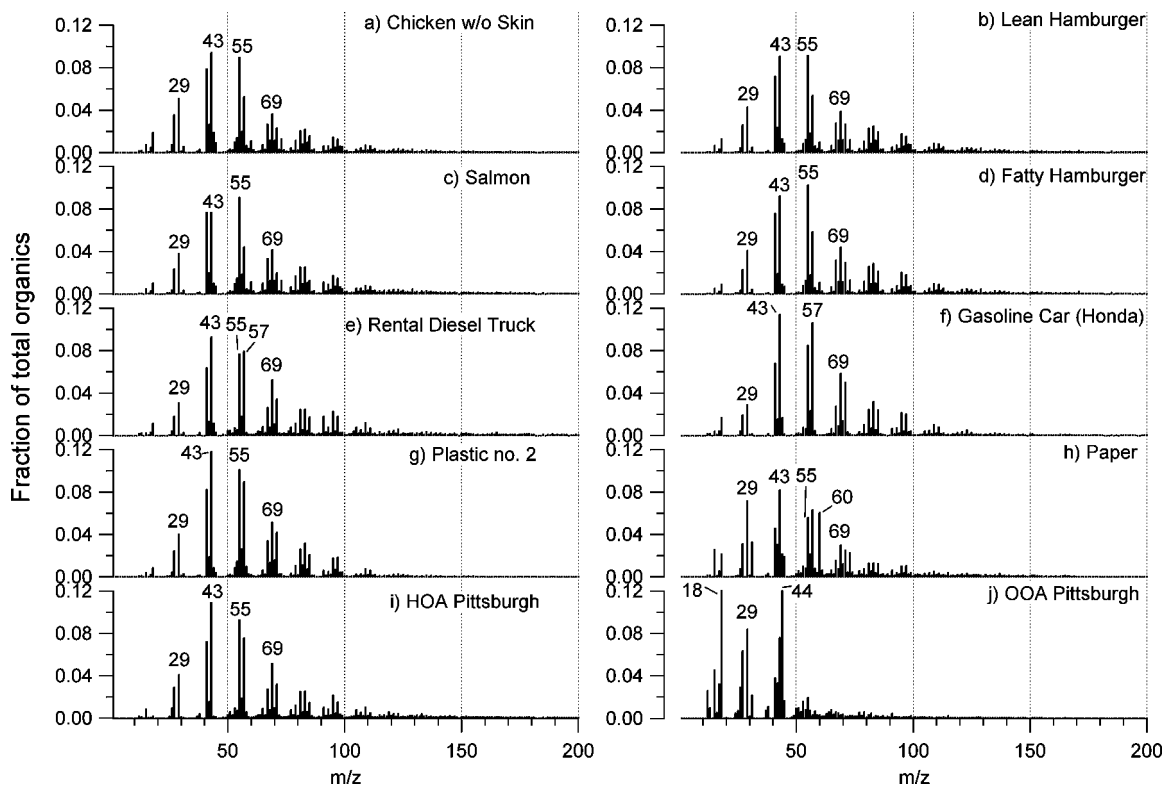
FIGURE 1. UMR mass spectra from selected POA sources. Also shown are HOA and OOA spectra from Pittsburgh (*7*).

Comparison with Database MS. We compared the UMR spectra of all experiments from this study to each other and to UMR MS from the AMS database (*9*). Pearson's *r* was used as a measure of spectral similarity, as shown in Figure 2. We also calculated *r* including only *m/z* > 44, because the few large intensities of some *m/z* < 44 can influence *r* disproportionately. However, we did not observe significantly different values or patterns, and only Pearson's *r* using the whole spectrum is used here. As discussed earlier the organic MS from this study are all broadly similar to each other (except for plastic no. 6) and thus the pattern of the similarities with MS from the database is quite uniform. Generally, the MS from meats, motors, and plastic burning are similar to those from HOA or POA (*r* > 0.8) and different from SOA or OOA MS (*r* < 0.6). The paper burning MS is most similar to those from biomass burning (*r* = 0.78 and 0.82, respectively), a finding consistent with the high *m/z* 60 in the paper spectrum. Even though the OA emissions from meat cooking are dominated by oxygenated fatty acids, their MS are not similar to ambient OOA spectra, but are more similar to spectra from HOA, such as those from traffic emissions.

Elemental Ratios. SI Table SI-5 shows elemental O/C and organic matter to organic carbon (OM/OC) ratios for several sources calculated with the method of Aiken et al. (*23, 24*). The vehicle and plastic OA emissions which are dominated by hydrocarbon-like ions have the lowest O/C ratios (0.03−0.04). The slightly higher degree of oxygenation of fatty acids (e.g., oleic acid $C_{18}H_{34}O_2$, with an atomic O/C ratio of 0.11) results in O/C ratios in the range 0.11−0.14 for the meat cooking OA, about 4 times larger than the vehicle ratios. All of these sources have O/C ratios far lower than ambient OOAs (O/C ∼ 0.5−1) (*24*). O/C from paper burning (0.31) is the highest for the three source groups while still lower than for ambient OOA, highlighting the importance of species such as levoglucosan, containing abundant oxygen, for paper smoke. This value is the same as for fresh biomass burning observed in the field (*24*). OM/OC ratios range from 1.22 (diesel vehicle) to 1.59 (paper).

Tracer Fragments. Some AMS UMR ions such as *m/z*s 43, 44, 57, and 60 are typically used as tracers of total organics, OOA, HOA, and BBOA, respectively, since they often correlate strongly with these OA components from statistical analysis of ambient data (*6, 7, 22*). It is of great interest to examine the patterns of these tracer fragments in the MS of sources studied here and to compare with ambient and chamber SOA MS in order to assess possible interferences and/or uniqueness of these tracers when analyzing a complex mixture of sources in urban air. Figure 3 shows ratios of these UMR *m/z*s to total organics (hereafter as, e.g., "44/Org"), whereas Figure 4 shows the high resolution spectra of these ions. Also shown are data from recent campaigns in Riverside, California (SOAR-1, July−August 2005) (*25*), and Mexico City (MILAGRO, March 2006) (*24*). The OA in Riverside, an inland location on the highly polluted Los Angeles basin, was dominated by SOA, especially in the afternoons, with maximum POA influence during the morning rush hour (*25*). The OA in Mexico City showed a similar diurnal cycle but was also influenced at times by large biomass burning OA (BBOA) plumes. PMF analysis of the Mexico City data identified strong OOA, HOA, and BBOA components. Spectra from time periods dominated by different types of OA, as well as PMF component spectra are also shown in Figures 3 and 4 for comparison with the source spectra. Finally, spectra from traditional SOA chamber experiments representative of biogenic and anthropogenic precursors (*24*) are also shown.

As a fraction of total organic signal, 43/Org shows the least variation across the sources and ambient data, consistent with previous observations of the correlation of this fragment with the total organic concentration and the lack of source differentiation from its UMR signal. As shown in Figure 4a−f, two ions contribute to UMR *m/z* 43: $C_3H_7^+$ and $C_2H_3O^+$, and as the latter increases for more oxygenated species, the former decreases creating a relatively constant fraction of UMR organic spectra. $C_3H_7^+$ is larger for all meat cooking experiments, indicative of the lower degree of
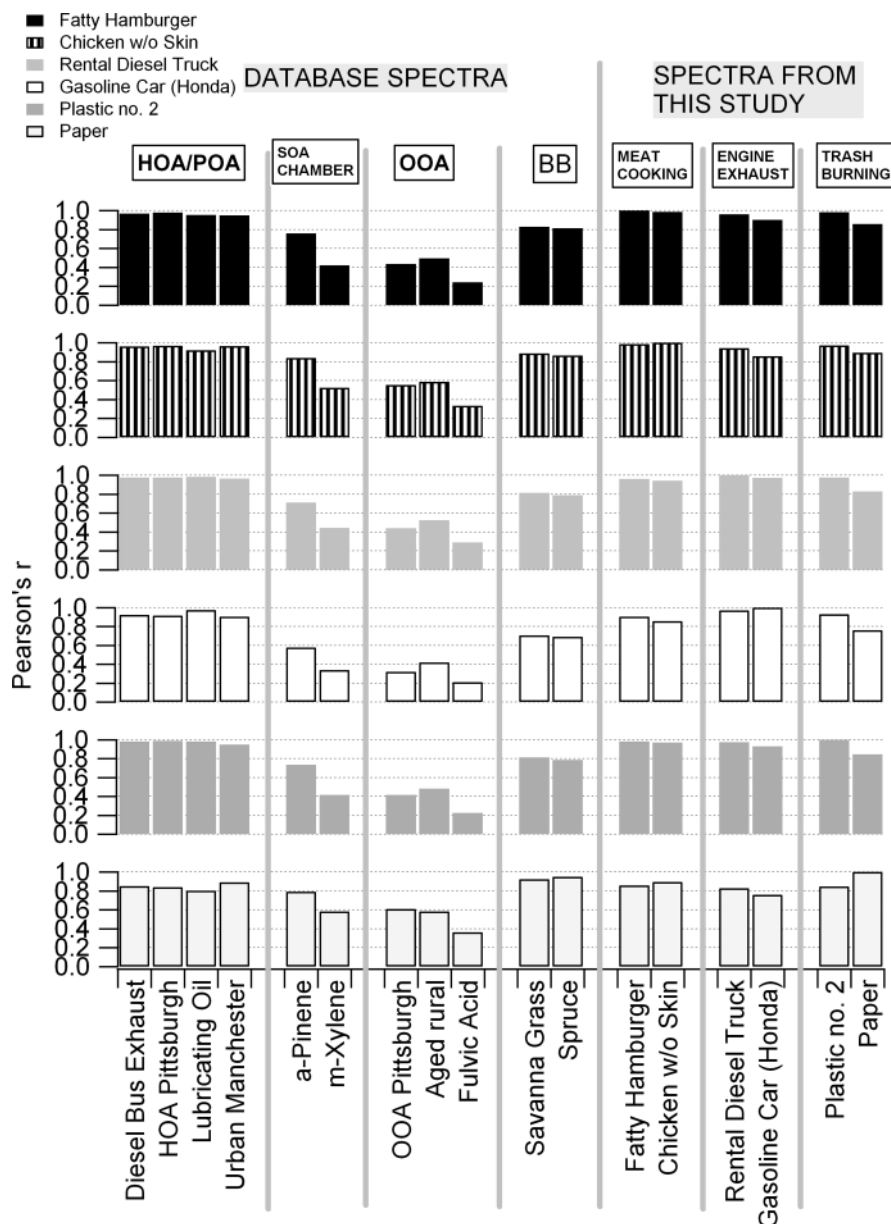
FIGURE 2. Pearson's *r* of UMR mass spectra from several POA sources studied here with spectra from sources and factor analysis.

oxygenation of these compounds as discussed above. In engine exhausts m/z 43 is completely dominated by $C_3H_7^+$ again supporting the dominant contribution of hydrocarbons to engine exhaust. Particles from burning plastic are also dominated by $C_3H_7^+$, whereas paper burning produces signals of similar intensity at $C_3H_7^+$ and $C_2H_3O^+$, consistent with its higher oxygen content. Ambient spectra during high OOA/SOA periods show a dominance of $C_2H_3O^+$ (consistent with chamber SOA spectra), whereas $C_3H_7^+$ is more important during rush hour periods and BB plumes. These patterns observed at m/z 43 are repeated at other low m/z ions (not shown), and are indicative of the ability to determine the degree of oxygenation of the particles with the HR-ToF-AMS (*23*).

m/z 44 is a characteristic fragment of oxidized organic compounds and is small when sampling primary aerosol materials such as hydrocarbons. Previously reported values of 44/Org typically range between 10 and 20% for ambient OOA (*7*). For all POA source groups studied here 44/Org ranges from about 0.5 to 2.5%, which is very low compared to most ambient total OA and especially ambient OOA spectra, as well as chamber SOA. This again confirms that

aerosol emitted from the sources studied here would not be misinterpreted as OOA/SOA in ambient air. Meats have 44/Org ~1–2%, with the fatty meats at the lower end and the leaner meats at the upper end. Ambient periods dominated by HOA or BBOA (as determined by PMF) and the HOA and BBOA PMF factors have low levels of 44/Org which are consistent with the POA sources reported here. Five ions compose most of the signal at m/z 44 (Figure 4g): CO_2^+, $C_2H_4O^+$, $C_3H_8^+$, and the ^{13}C isotopes of $C_2H_3O^+$ ($^{13}C^{12}CH_3O^+$) and $C_3H_7^+$ ($^{13}C^{12}C_2H_7^+$). $C_2H_4O^+$ and $^{13}C^{12}CH_3O^+$ are not well separated at the resolution of this spectrometer and appear together in between the other two ions, and similarly for $^{13}C^{12}C_2H_7^+$ and $C_3H_8^+$. Thus the three groups of ions at m/z 44 represent (from left to right) highly oxygenated, oxygenated, and reduced molecular fragments, respectively. Except for pork ribs, whose m/z 44 is dominated by $C_2H_4O^+$, all meat OAs show similar contributions of all three ions. The 44/Org ratio for engine exhaust is about 1%. The same three ions are observed with slightly different patterns, depending on the specific source. Plastic burning also produces low 44/Org levels, with similar contributions of all ions. Paper burning has the highest percentage of 44/Org of all sources
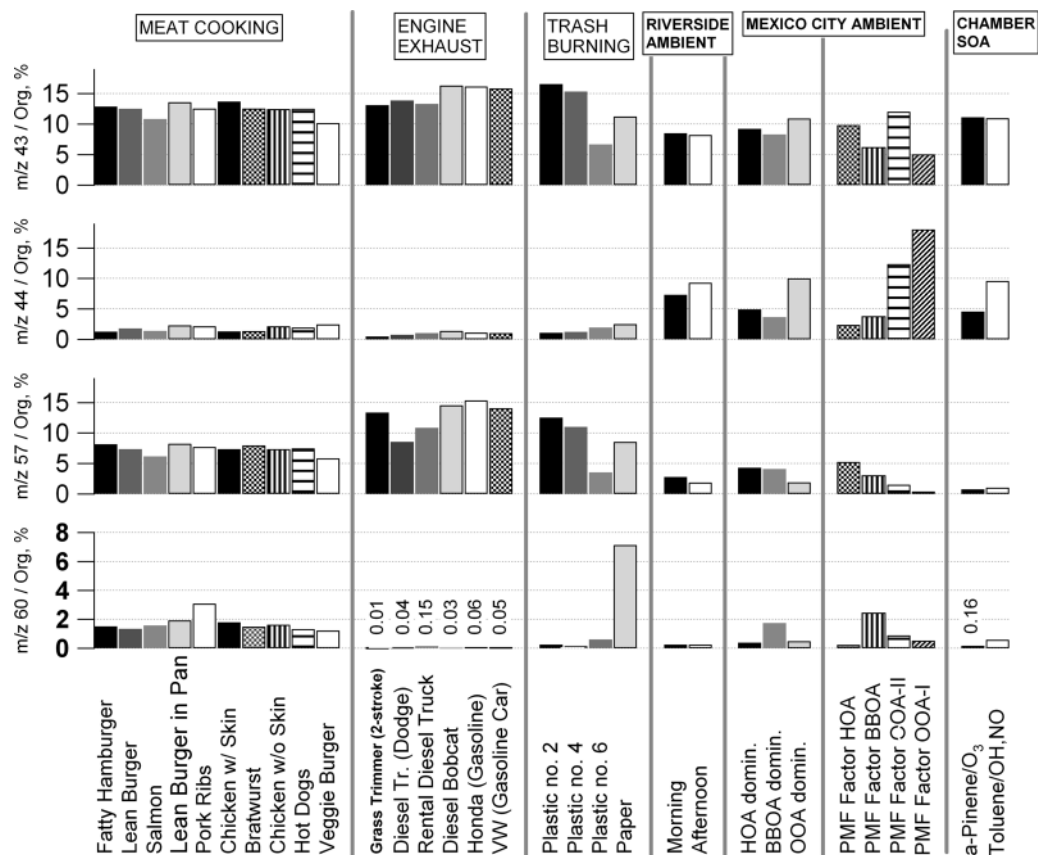
FIGURE 3. Ratios of m/z 43, 44, 57, and 60 to organics for the three source groups and comparison to same ratios for ambient organic aerosol from Riverside (CA), Mexico City (ambient and PMF factors), and chamber SOA.
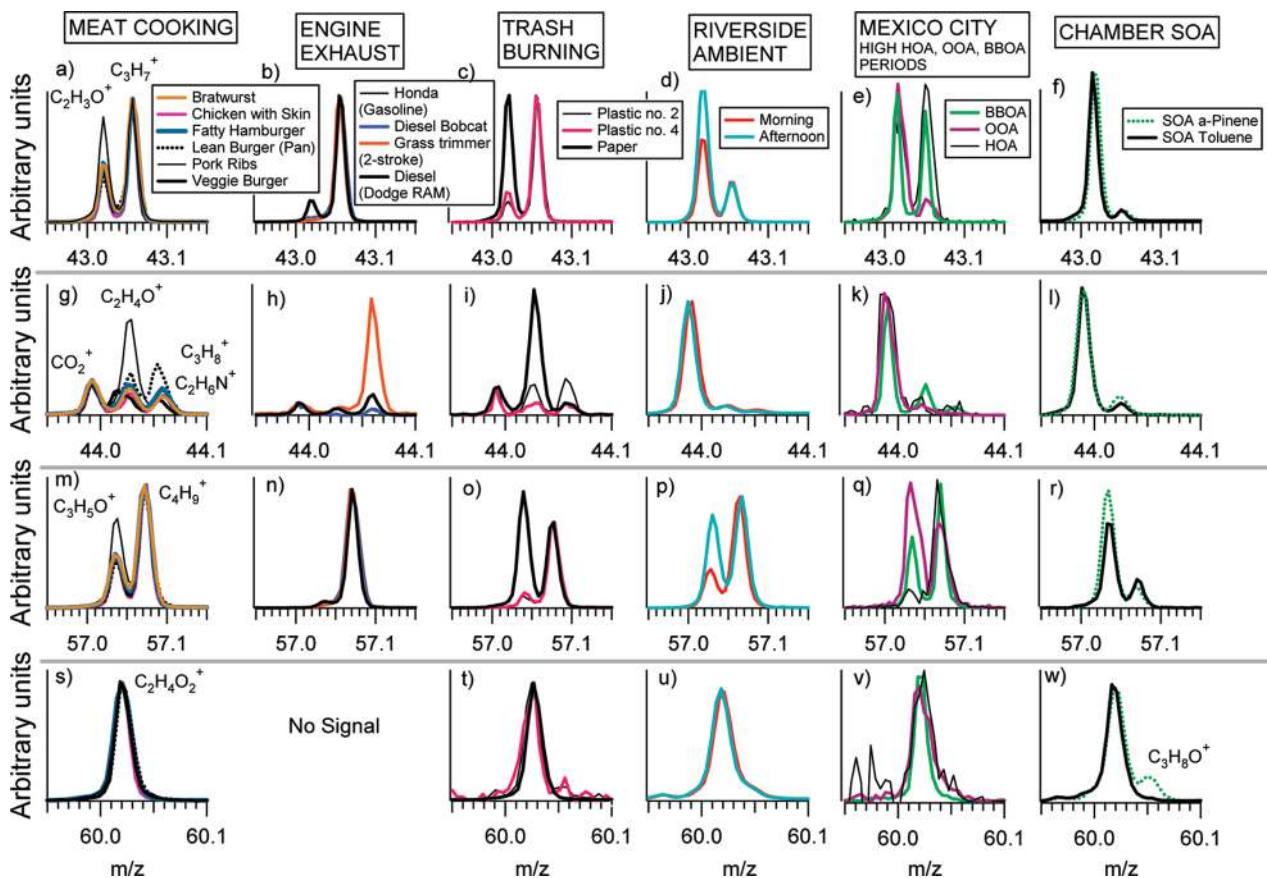

FIGURE 4. High resolution ion signals at m/zs 43 (a–f), 44 (g–l), 57 (m–r), and 60 (s–w).

measured (about 2.5%, but still much lower than ambient OOA), indicating that this source has a larger contribution from oxidized compounds. This signal is dominated by $C_2H_4O^+$, contrary to the CO_2^+ dominance typically observed in ambient air. In chamber SOA and ambient OA, however, the CO_2^+ ion dominates in all cases, highlighting the fact that a large POA contribution from the primary sources studied here can easily be distinguished from ambient OOA/SOA using HR-ToF-AMS data. Ambient periods with high BBOA are dominated by CO_2^+, in contrast to the paper burning spectra. However 44/Org is low during high BBOA periods, so the high CO_2^+ may instead be associated with the OOA which is still present during those periods. Alternatively, ambient BBOA emissions or aging may lead to more oxidation than for the paper burning OA studied here.

UMR 57/Org has been used in several studies as an indicator of saturated hydrocarbon compounds ($C_4H_9^+$) and can include some contribution from oxidized organic compounds due to $C_3H_5O^+$ (6, 10). Figure 3 shows that 57/Org is high for all the POA sources studied here, consistent with the association found in previous studies. This value is ~6–8% for meats, 8–15% for motor vehicles, and ~8–12% for trash burning, except for polystyrene at ~3%. An analysis of the contributions of specific ions to m/z 57 (Figure 4m–r) shows that for all sources the contributions of oxygenated vs reduced fragments are similar to those at m/z 43. Engine exhaust is overwhelmingly dominated by $C_4H_9^+$, which again supports the good correlation of this ion with urban sources of CO and NO_x which are strongly influenced by traffic (6, 26, 27). Ambient OOA-dominated air, the OOA factors from PMF, and chamber SOA all have low m/z 57/Org (~1–2.5%). The source and ambient spectra at m/z 57 again support the conclusion that the POA sources studied here do not make a direct significant contribution to ambient OOA.

UMR m/z 60 is a marker for biomass burning (22). A closer look at the high resolution signal (see Figure 4s–w) shows that only one ion ($C_2H_4O_2^+$) contributes to the signal in almost all spectra analyzed here. COS^+, which may arise from organosulfates, is detectable in the Riverside spectra, indicating a possible small concentration of these species, whereas $C_3H_8O^+$ is visible in the α-pinene SOA spectrum. For the meat OAs 60/Org has values ~1.5 to 3%, whereas typical values for SOA or OOA are ~0.2–0.4%. This implies that excess m/z 60 above ~0.4% is not an exclusive tracer of biomass (or paper) burning in ambient air, as some of its signal may arise from meat cooking. For all motor vehicle sources 60/Org is 0.15% or lower and averages ~0.07%. This suggests that significant signal at m/z 60 in ambient air indicates that sources other than vehicle exhaust are important contributors to aerosol concentrations. As for the trash burning sources, plastics show very small fractions of m/z 60 (<1%). Paper, however, has a large contribution from mass fragment 60 to total organics (~7%), which is as high as the contribution from mass fragments 43 and 57. This finding indicates that "biomass burning" components in ambient data may have contributions from trash burning.

Implications for the Interpretation of AMS Factor Analyses. Our results strongly suggest that none of the sources analyzed here are an important source of the ambient OOA identified in AMS factor analysis studies. This reinforces the conclusion from previous studies (1, 6–9) that the main source of ambient OOA is atmospheric oxidation, mostly as SOA from various precursors (anthropogenic, biogenic, and biomass burning) but perhaps also with some contribution from heterogeneous oxidation. Motor vehicles, plastic burning, and meat cooking are very likely to be retrieved as a single component in AMS UMR factor analysis due to the similarity of their spectra, as previously suggested (6, 7). Paper burning will likely be retrieved together with ambient BBOA. Factor analysis of HR data may allow better separation of meat cooking from the other sources due to the larger differences in HR spectra. Since only a limited number of sources and conditions were analyzed here, additional experiments will be useful to confirm the generality of these results.

Supporting Information Available
Additional experimental details including supplementary figures and tables. This material is available free of charge via the Internet at http://pubs.acs.org.

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1 # Supplementary Information to

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3 **Characterization of Primary Organic Aerosol Emissions from Meat Cooking, Trash Burning,**

4 **and Motor Vehicles with High-Resolution Aerosol Mass Spectrometry and Comparison with**

5 **Ambient and Chamber Observations**

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20 Number of pages: 10

21 Figures: SI-1 – SI-3

22 Tables: SI-1 – SI-5

Additional Experimental Details

The CO2 emissions were measured with a LI-6262 CO2/H2O Gas Analyzer (LI-COR, Lincoln, NE).

A DustTrak portable laser photometer (Model 8520, TSI, Shoreview, MN) was also operated continuously with a PM1 impactor inlet. It measures the light scattered at 90 degrees, which is roughly proportional to the aerosol mass concentration as long as the size distribution and refractive index are approximately constant *(1, 2)*. A calibration factor of 0.3 was used to estimate PM_1 mass from the DustTrak. This factor was determined in a previous study *(2)*, and is smaller than 1 as expected from the higher scattering efficiency of submicron particles compared to dust.

An AMS collection efficiency of 1 was used for all experiments since these sources were expected to produce liquid organic particles which are collected with high efficiency in the AMS. Variations in the collection efficiency between experiments could cause some additional uncertainty, which is likely small compared to other sources of uncertainty for the emission ratios determined here.

Emission Ratio Calculation

Emission ratios of PM to CO_2 were determined for all sources and are shown in Tables SI-2-4 and Figure SI-2 only to provide context for these experiments, and for comparison with the conditions of future experiments performed by other groups. These ratios should not be used to estimate emissions from these types of sources since the experimental setup was not designed for accurate quantification.

For calculating emission ratios of the various sources the AMS data (particle concentrations in µg m^{-3}) were processed with standard methods *(3)* and normalized to the enhancement in CO_2 concentrations (in ppm) between the beginning of the experiment and the point measurement. Note that the origin of the CO_2 is different in the different experiments: fuel combustion in the mobile sources, material combustion in the trash burning experiments, and the fuel burned by the cooking grill for the meat cooking experiments. Thus the comparison of emissions ratios between different source groups can only be qualitative. An effort was made to operate the grill in a consistent manner so that the amount of CO_2 emitted by the grill when cooking each piece of meat was of the same order, which allows to self-consistently compare the meat emission ratios from this study.

Tables

Table SI-1: Order-of-magnitude estimate of meat cooking aerosol concentrations for the Greater Los Angeles (GLA) area (~12,500 km^2)

Quantity	Value
Consumed meat per person (lbs / yr) *(4)*	200 lbs / year
Consumed meat per person (kg / day)	0.25 kg / day
PM emitted per kg of meat cooked	10 g PM / kg meat
Population of GLA *(5)*	1.8×10^7
Meat consumed in GLA	4.4×10^6 kg / day
Meat cooking PM emitted in GLA	4.4×10^4 kg / day
Footprint of GLA basin	12500 km^2
Average boundary layer height	1500 m
Estimated air volume of GLA basin	1.875×10^{13} m^3
Assumed residence time of emissions	1 day
Average concentration of Meat Cooking PM in GLA area	**2.3 µg m^{-3}**

55   **Table SI-2:** Meat sources used in this work, their fat contents, grouping into fat groups according to

56   fat content, and their emission ratios. Veggie burgers are included in the meat source group as they

57   are an increasingly common meat substitute.

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Meat	Meat Specifications	Fat content (%)	Fat Group	Weight (lbs)	Emission Ratios (OA/ excess CO_2, in µg m^{-3}/ppm)
Veggie Burgers	Boca Meatless Burgers Originally Vegan	0.7	1	0.63	0.3
Chicken without Skin	RedBird All Natural Boneless, Skinless Chicken Breast with Rib Meat	1.8	1	1.025	1.1
Chicken with Skin	RedBird All Natural Chicken Split Breast	4.5	2	1.095	5.0
Salmon	King Soopers Wild King Salmon Fillet, previously frozen	6.3	2	1.08	9.00
Lean Burger	King Soopers Ground Beef Patties with Natural Flavor	9	2	1.35	12.5 (grill) 7.5 (pan)
Fatty Hamburger	King Soopers Ground Beef Patties with Natural Flavor	20	3	1.35	37.2
Bratwurst	Boulder Sausage Bratwurst, made with Natural Ingredients	20	3	1	3.4
Pork Ribs	Kroger Moist & Tender Silverplatter Pork Loin Backribs	23.8	3	2.96	7.2
Hot dogs	Oscar Meyer Wieners made with Turkey, Pork and Chicken	26.7	3	0.5	0.2

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60 **Table SI-3:** Mobile sources tested in this work and their emission ratios
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Mobile Source	Specifications	Emission Ratios (OA/ excess CO_2, in $\mu g\ m^{-3}$/ppm)
Gasoline Car 1 (Honda)	Honda Accord, 1995, 4-cylinder, 85-octane gasoline	0.016
Gasoline Car 2 (VW)	VW Golf, 1996	0.019
Diesel Truck 1	Dodge RAM 250 pickup truck, 1993, Diesel	3.5
Diesel Truck 2 (Penske Rental Truck)	16 inch Diesel Cargo Van	0.91
Grass trimmer (2-stroke gasoline)	Shindaiwa Trimmer and Brushcutter, 2-stroke engine	18.7
Small Diesel Tractor (Skid Steer Loader, Bobcat brand)	Diesel, Model 733, 1980	0.11

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63   **Table SI-4:** „Trash" burning sources tested in this work and their emission ratios

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Trash Material	Specifications	Emission Ratios (OA/ excess CO_2, in µg m^{-3}/ppm)
Plastic no. 2	Supermarket Bags; high-density Polyethylene	4.5
Plastic no. 6	Cup, Polystyrene	0.39
Plastic no. 4	Bags, low-density Polyethylene	1.46
Paper	Brown Supermarket Paper Bags	4.2

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66   **Table SI-5:** Elemental Ratios for OA source groups meat cooking (fatty hamburger, chicken without

67   skin), trash burning (plastic no. 2, paper), and combustion engines (diesel, gasoline vehicle)

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| Sample	O/C	OM/OC
Fatty Hamburger	0.11	1.32
Chicken without Skin	0.14	1.36
Plastic no. 2	0.08	1.29
Paper	0.31	1.59
Diesel Vehicle	0.03	1.22
Gasoline Vehicle	0.04	1.24

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70　**Figures**
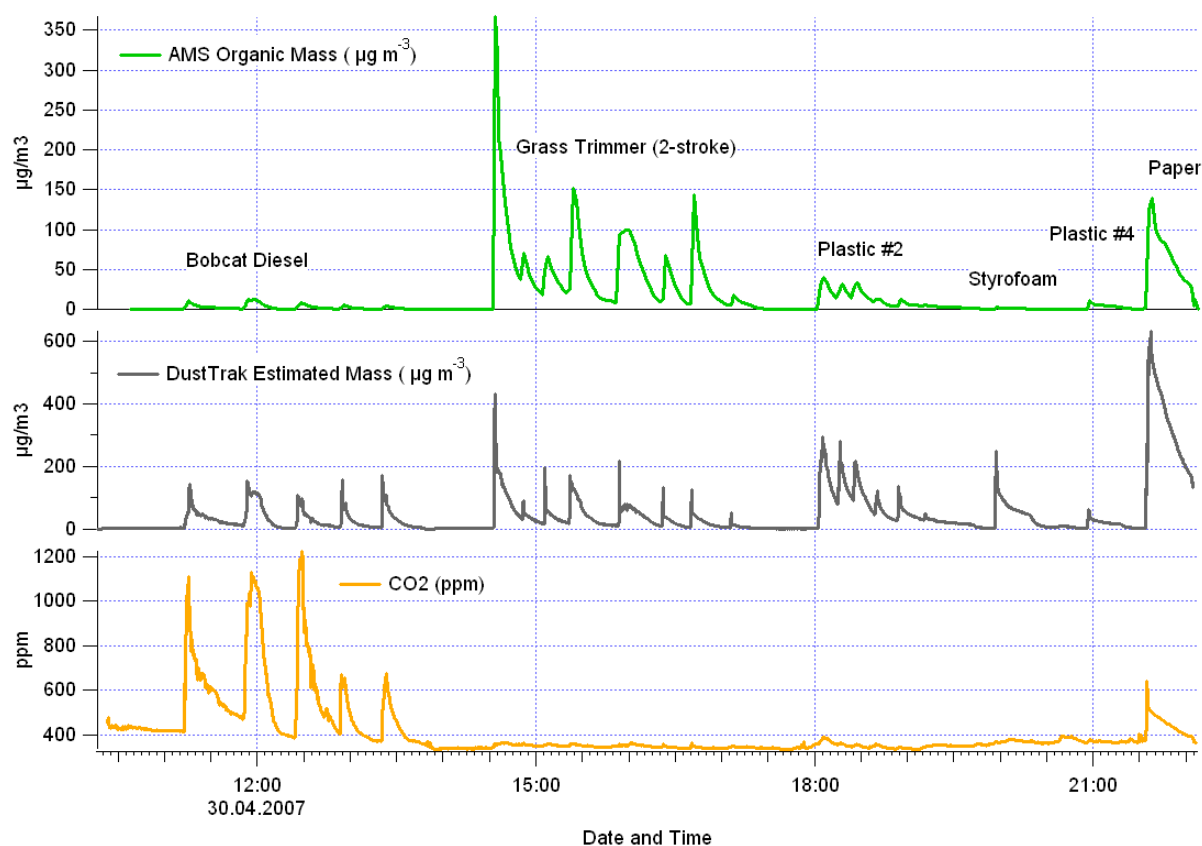
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73　**Figure SI-1:** Time traces of OA measured by the AMS, PM_1 estimated from the DustTrak, and of

74　CO_2 for source testing experiments made on 4/30/2007. Individual spikes in the time series indicate

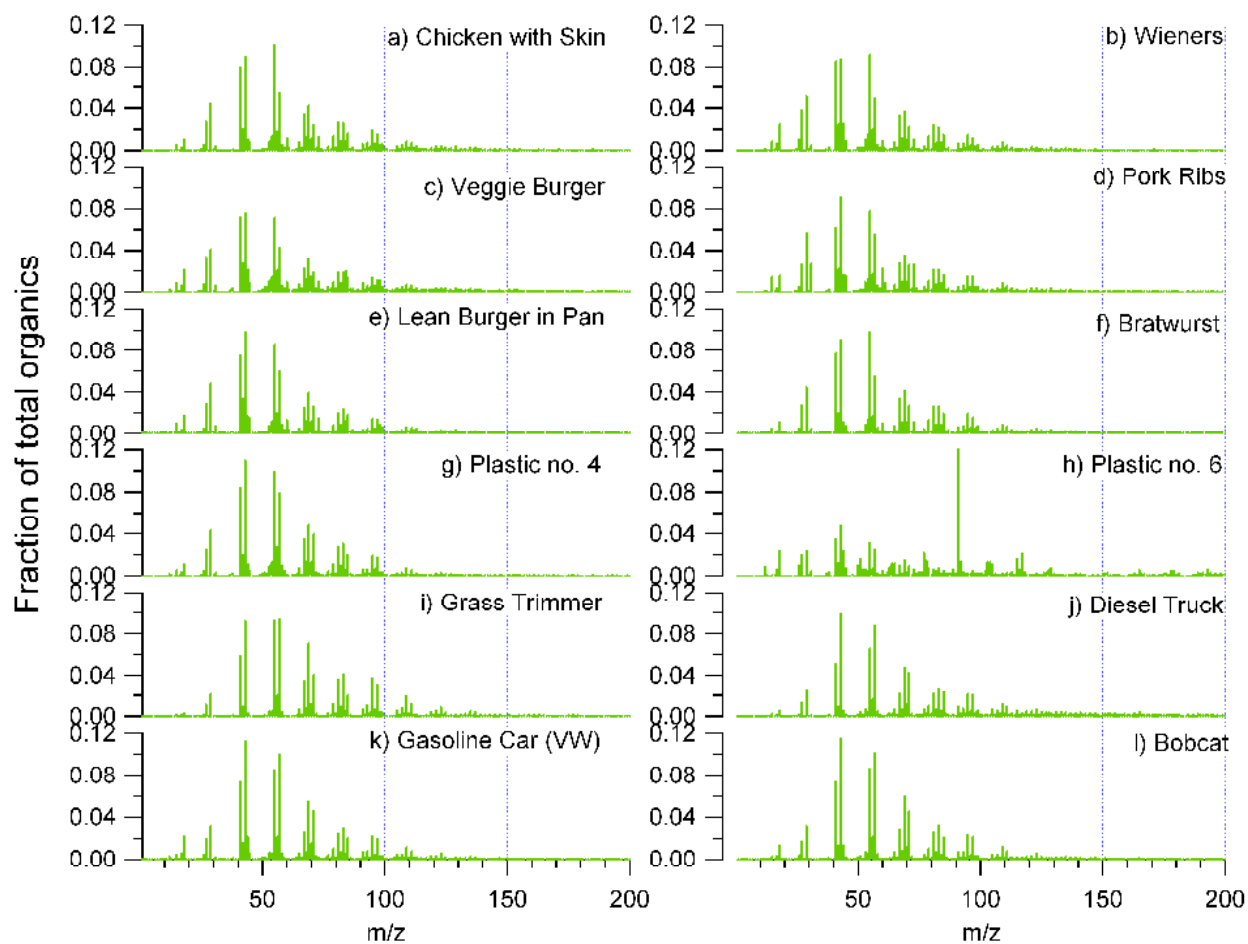75　consecutive starts of burns or engines. See text for description.

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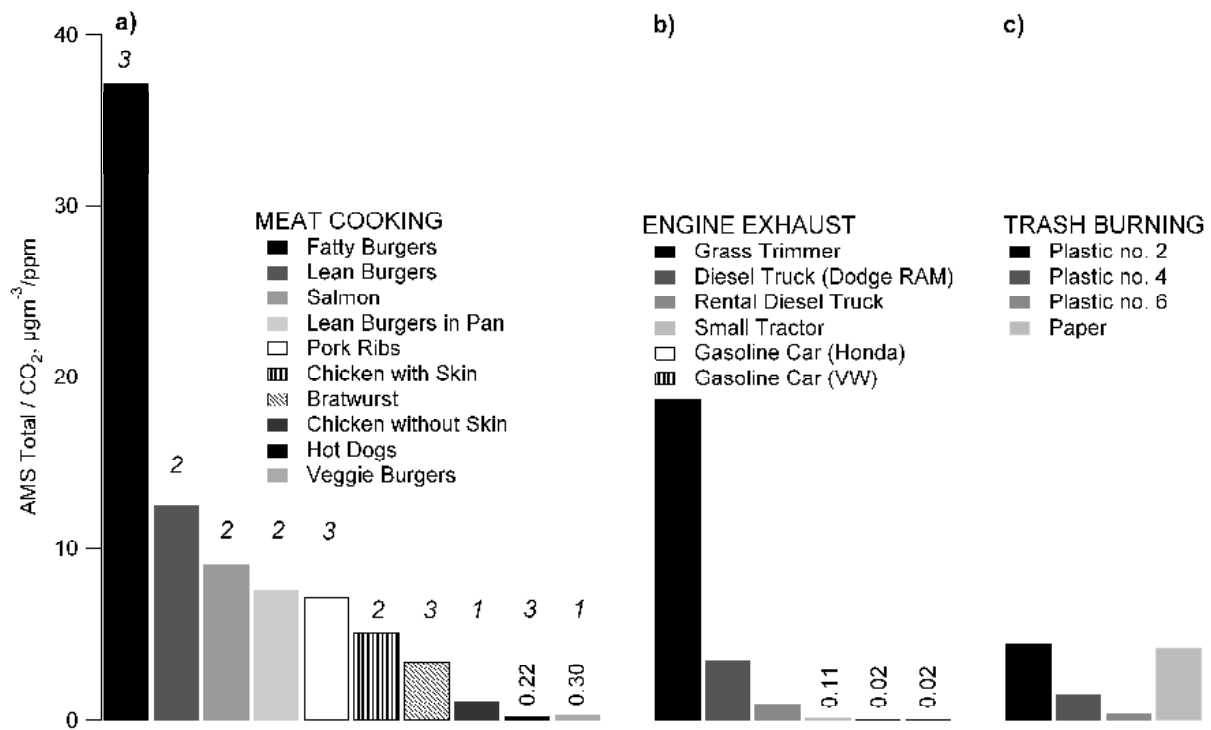
Figure SI-2: Emission ratios (particle concentration enhancements normalized to CO_2 enhancements, µg m^{-3}/ppm) for the following source groups: meat cooking (a), engine exhaust (b), trash burning (c). Numbers in italics refer to meat fat groups (see Table SI-2).

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Figure SI-3: Mass spectra (unit mass resolution) from meat cooking experiments (a-f), trash burning

experiments (g-h), and engine combustion (i-l).

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References Supplementary Info

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