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Inferring Aerosol Sources from Low-Cost Air Quality Sensor Measurements: A Case Study in Delhi, India

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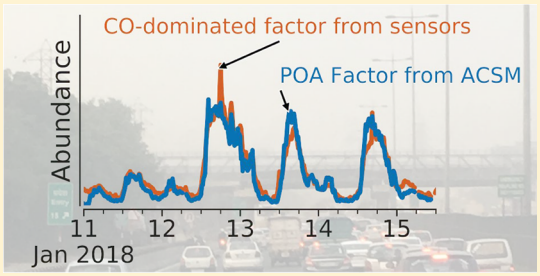
Supporting Information

ABSTRACT: Low-cost sensors (LCS) offer the opportunity to measure urban air quality at a spatiotemporal scale that is finer than what is currently practical with expensive research- or regulatory-grade instruments. Recently, the LCS research community has focused largely on sensor calibration, pollution monitoring, and exposure assessment; here, we investigate the applicability of LCS for characterizing particulate pollution sources in an urban environment. Using an integrated multipollutant LCS system (which measures both gases and particles), we collected air quality data for 6 weeks during the winter at a site in Delhi, India. The results were compared to measurements taken by co-located research-grade particle instruments. Non-negative matrix factorization was used to deconvolve LCS data into unique factors that were then identified by examining the factor composition and comparing them to the research-grade measurements. The data were described well by three factors: a combustion factor characterized by high CO levels and two factors characterized by measured particles. These factors align well with measurements by research-grade instruments, including particle types determined from factor analysis of online particle composition measurements. This work demonstrates that multipollutant LCS measurements, despite their inherent limitations (e.g., calibration challenges and inability to measure smallest particles), can provide insight into sources of fine particulate matter in a complex urban environment.



1. INTRODUCTION

Measurements of air pollution are crucial for managing regional air quality, evaluating existing policy, and estimating human exposure to air pollutants. However, collecting air quality data using continuous air quality monitoring stations or research-grade atmospheric chemistry instrumentation can be very expensive, and hence such measurements tend to be quite limited in spatial resolution and/or are only gathered during relatively short measurement periods. The rapid rise and adoption of low-cost air quality sensors (LCS) presents a new opportunity to dramatically increase the spatial and temporal resolution of existing air quality measurements, assess the effectiveness of air quality interventions, and better quantify human exposure to air pollution.[1] Until now, the focus has primarily been on assessing the accuracy and reliability of new sensor technologies and calibration approaches[2−8] and the routine monitoring of air quality.[9−11] There has been relatively little work exploring applications of LCS for improving our understanding of the underlying factors that control air pollution levels, such as identifying sources of pollutants.[12]

Here, we demonstrate the use of LCS to better understand the types and sources of atmospheric aerosol particles in a complex urban environment. Measurements are taken in Delhi, India, a megacity whose high levels of air pollution have negative impacts on health[13−17] and the economy.[14] Pollution sources, which can include crop burning, biomass burning, vehicle use, and industrial emissions,[18−21] are poorly constrained and exhibit high spatial and temporal heterogeneity. Central to this work is the use of multipollutant sensors that measure both gases and particles as well as the co-location of these sensors with several research-grade instruments that measure particle size and composition. The information from these instruments, which have been collecting data for more than one year on the composition and sources of PM in Delhi, is leveraged to provide information about how LCS can be

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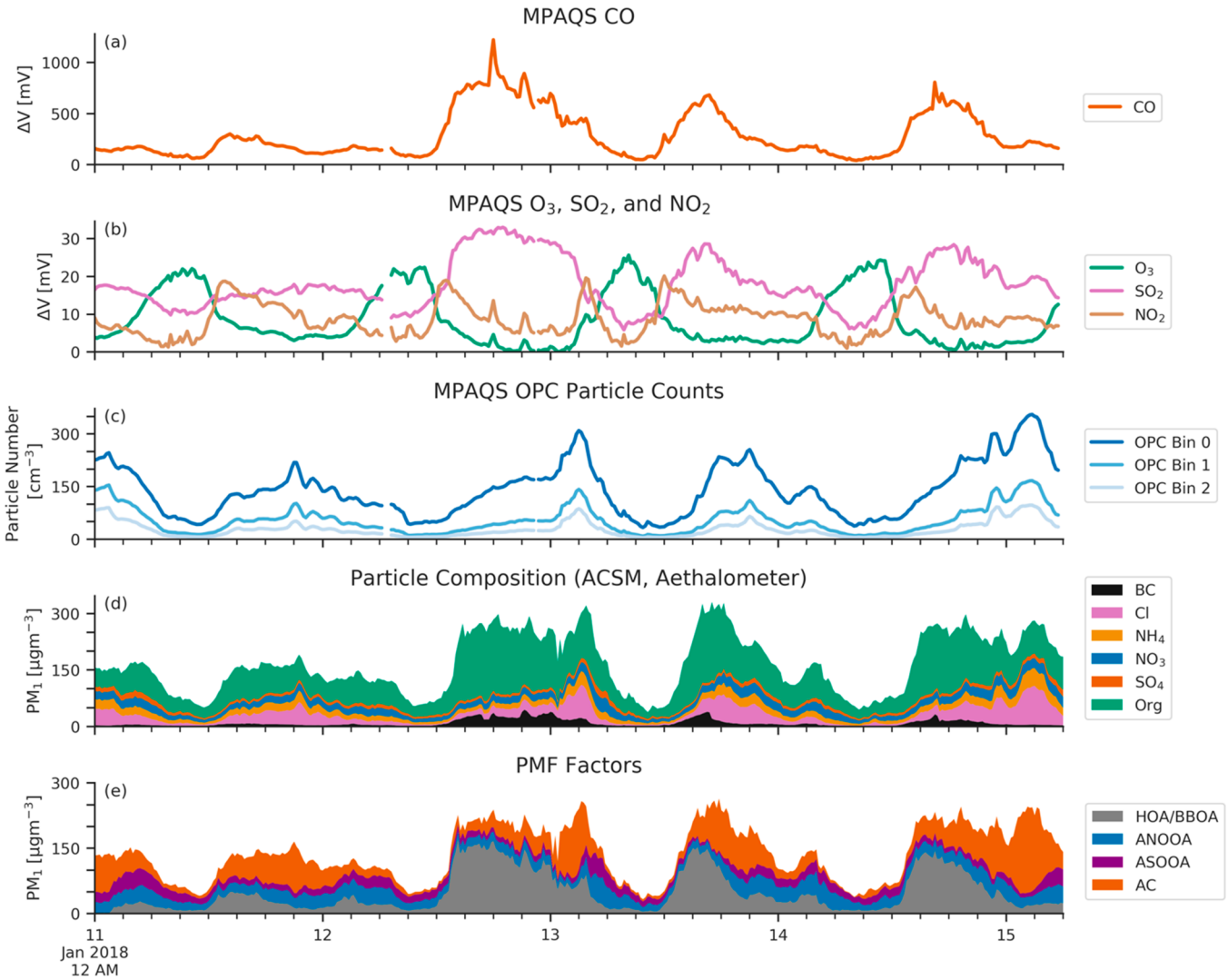

Figure 1. Time series for the first 4 days of deployment, showing measurements taken by the MPAQS (a−c), particle composition measurements taken using an ACSM and an Aethalometer (d), and positive matrix factorization (PMF) results from the ACSM data (e). Factors obtained from combined organic/inorganic PMF analysis (described by Bhandari et al.[21]) include a factor for primary organic aerosol [mixed hydrocarbon-like and biomass burning organic aerosol (HOA/BBOA)], a factor for ammonium nitrate and oxygenated organic aerosol (ANOOA), a factor for ammonium sulfate and oxygenated organic aerosol (ASOOA), and a factor for ammonium chloride (AC). OPC bins 0−2 nominally correspond to particles with sizes of 0.38−0.54, 0.54−0.78, and 0.78−1.05 μm, respectively, though sizes may not be exact due to differences namely in RH and particle optical properties (i.e., refractive index).

used to better constrain the concentrations, variability, and sources of air pollution in highly polluted areas.

2. MATERIALS AND METHODS

Measurements were taken at the Delhi Aerosol Supersite,[22] where research-grade instrumentation has been sampling nearly continuously since January 2017. Measurements were taken from a laboratory on the IIT-Delhi campus (Hauz Khas, South Delhi), located on the fourth floor (∼20 m) of the building and ∼150 m from a major arterial roadway. Here, we focus our analysis on January 1, 2018, to February 14, 2018, the time period for which data were available across all instruments. Research-grade instruments included an aerosol chemical speciation monitor (ACSM, Aerodyne Research, Inc.) measuring real-time non-refractory PM_1 (NR-PM_1) composition, an Aethalometer (Magee Scientific) measuring black carbon (BC), and a scanning mobility particle sizer with a water-based condensation particle counter (SMPS, TSI Inc.) measuring the particle size distribution for particles with electrical mobility diameters between 12 and 560 nm. The PM_1 mass concentration is estimated by fitting the volume-weighted particle size distribution below 560 nm and extrapolating to 1 μm; this approach is detailed in the Supporting Information (section 8), and results compare well with the total ("C-PM_1") mass used by Gani et al.[22] The results from each of these instruments and a description of the measurement site and protocols are provided elsewhere.[21,22]

LCS measurements were taken using a custom-designed sensor node [multipollutant air quality sensor (MPAQS)] for continuous, real-time measurements of several gas-phase pollutants (SO_2, NO_2, O_3, and CO), particulate matter, and environmental variables (relative humidity and temperature). Gas-phase pollutants were measured using four-electrode electrochemical sensors (Alphasense SO2-A4, OX-A431, NO2-A43F, and CO-A4) with the Alphasense analog potentiostat circuitry. These sensors have been described in

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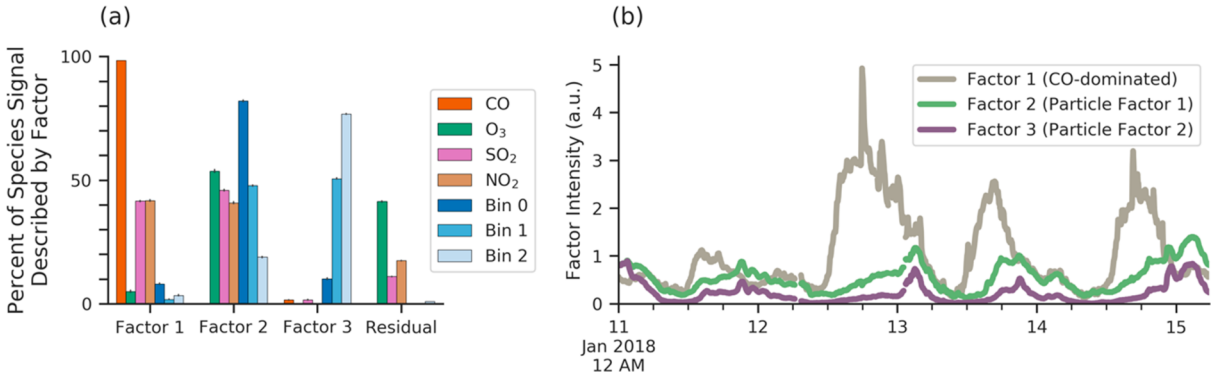

Figure 2. Three-factor NMF solution based on gas- and particle-phase inputs from the MPAQS unit. (a) Fraction of each input species associated with a given factor. For example, 95% of the CO signal belongs with factor 1, along with less just under half of the SO_2 and NO_2 signals, and small fractions of the particles. Factor 2 is associated mostly with the particles in bins 0 and 1 (nominally the smallest ones measured). The third factor is dominated by the bin 2 particles, with progressively smaller contributions from bins 1 and 0. Confidence intervals (95%, determined from the bootstrapping procedure) are shown but are generally under 0.5% and so are not clearly visible. (b) Four-day snapshot of the time series trace for each factor, shown in arbitrary units (a.u.).

detail previously[23] and so are not described in detail here. The OX-A431 sensor measures O_x (O_3 + NO_2), so O_3 is determined by subtracting the NO_2 sensor signal from the O_x sensor signal. Particles are measured using a modified commercially available optical particle counter (Alphasense OPC-N2) that uses a 658 nm laser to count and size particles into 16 size bins ranging from 380 nm to 17.5 μm. The OPC was modified by designing an airtight enclosure and replacing the fan with a pump, pulling at the same manufacturer-specified flow rate (350 SCCM). These modifications were made to accommodate a bifurcated flow diluter[24,25] upstream of the sensors to reduce the total particle loading moving through the MPAQS to prevent rapid degradation of the sensors. A differential pressure sensor was used to measure the pressure drop across the HEPA filter to correct for dilution. While particle measurements can be affected by water uptake,[26] the relative humidity (RH) within the sensor units was relatively constant (44 ± 10%), so RH changes are unlikely to affect the OPC measurements substantially. Additionally, particle sizing by OPCs can be strongly dependent on the refractive index of the particles;[26,27] rather than attempt to correct for this, we treat the OPC size bins simply as unique signals (from particles that may differ in both size and optical properties) and do not interpret the measurements in terms of true particle size distributions. More details regarding the MPAQS, and its expected response to RH changes, can be found in the Supporting Information (sections 1 and 13).

The MPAQS gas-phase data are used in their "raw" format, that is, as output voltages rather than mixing ratios. While recent work has shown that advanced nonparametric algorithms[2−4] enable the accurate calibration of electrochemical sensors, here we simply use the difference between the working and auxiliary electrodes (ΔV), which produces a value that is roughly proportional to the mixing ratio of the target analyte[23] and provides a first-order correction for effects related to environmental parameters (e.g., temperature and RH). This approach does not fully eliminate potential sources of error associated with electrochemical sensors, such as large or rapid temperature and RH changes[3,5−7] and cross-interferences with other chemical species,[4,28−30] but still provides a first-order metric for gas concentration. Moreover, by using only the raw data, we demonstrate that LCS can be used to infer pollution sources even without comprehensive characterization of the detailed sensor response to these confounding factors. Instead, the approach used here primarily requires that sensor output (in this case ΔV) be proportional to pollutant concentration.

Due to concerns related to potential sensor-to-sensor variability, three identical MPAQS instruments were deployed at the supersite (a fourth MPAQS experienced a pump failure almost immediately, and thus its data are excluded from this analysis). Here we show the results for one of the MPAQS units (SN015), though all three showed good agreement with one another; comparisons of the data from the three can be found in the Supporting Information (sections 2, 7, and 10).

3. RESULTS AND DISCUSSION

Figure 1 shows a four-day snapshot of both the LCS data (panels a−c) and data from the research-grade instruments (panels d and e). Panels a (CO) and b (O_3, NO_2, and SO_2) of Figure 1 show the ΔV values from the four MPAQS electrochemical sensors: O_3 exhibits a diurnal trend with a mid-day peak, while SO_2 and CO tend to peak in the evening. Figure 1c shows data from the three smallest size bins of the OPC (bins 0−2, chosen to align with PM sizes measured by the research-grade instruments); while they do co-vary during this time period, they can diverge at times, indicating a change in the perceived particle size distribution. Figure 1d shows the aerosol composition data from the ACSM and Aethalometer, showing a strong chloride signal, peaks of high black carbon, and consistently high organic loadings. Figure 1e shows the results of positive matrix factorization (PMF) analysis of ACSM data, as described by Bhandari et al.[21] Key features of these time series include: (1) the HOA/BBOA signal that appears to be strongest at night, when CO and SO_2 levels are highest, (2) ammonium chloride (AC) constitutes a major and sometimes dominant fraction of the PM_1 mass (discussed in detail by Gani et al.[22]), and (3) the levels of oxidized organic aerosol and associated inorganic salts (ANOOA and ASOOA) exhibit a slight diurnal behavior with peaks around early morning.[21]

To aid in the interpretation of the LCS data, we use non-negative matrix factorization[31−33] (NMF), an unsupervised learning approach that groups together species by covariance to obtain physically interpretable factors. NMF is similar to

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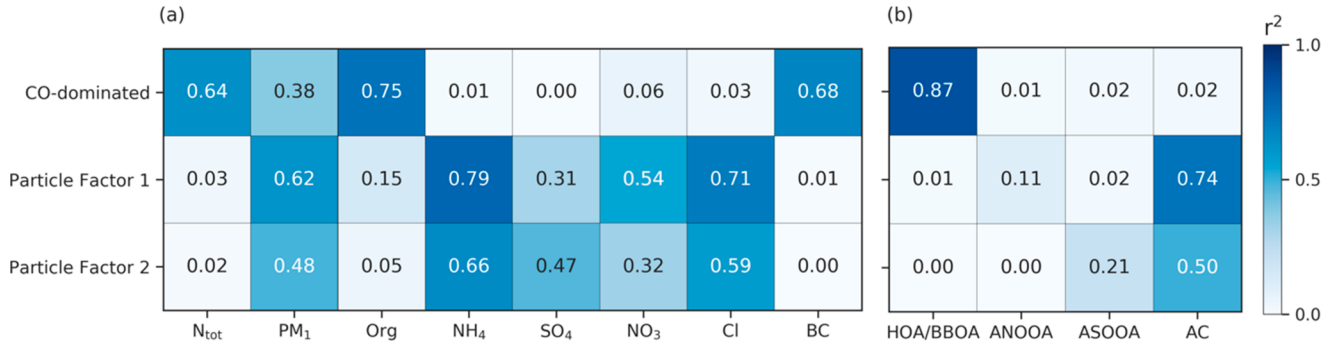

Figure 3. Correlations of the output of the low-cost sensor factor analysis to the physical and chemical properties of the aerosol, as measured by the co-located research-grade instrumentation. Numbers (and colors) in each cell denote the correlation coefficient (Pearson r^2) between the reference value (x axis) and NMF factor (y axis). (a) Correlation between the NMF results and the number concentration (N_{tot}), PM_1 mass concentration, and chemical composition (Org, NH_4, SO_4, NO_3, Cl, and BC) of the aerosol. (b) Correlation between each NMF factor and each ACSM PMF factor. Confidence intervals associated with all Pearson r^2 values, as determined from the bootstrap approach, are given in section 12 of the Supporting Information.

PMF, a technique commonly used in atmospheric aerosol chemistry[34,35] (though NMF has also seen recent use in the field[36]), with the main difference being how component scaling and error are treated. Specifically, NMF weights all components equally rather than by individual errors, which are currently not well-constrained for LCS. To estimate errors and validate the repeatability of our approach, we use a bootstrap approach[37] as documented in the Supporting Information (section 11). We combine data from both the gas and particle sensors into a single input matrix for the factorization, as has been done in a previous PMF study.[38] Use of seven input variables (O_3, SO_2, CO, NO_2, OPC bin 0, OPC bin 1, and OPC bin 2) from the MPAQS results in a $7 \times n$ input matrix, where n is the number of 1 min mean values from each sensor ($n = 61664$ for this analysis). We then use NMF to decompose the input matrix into factors with unique input ratios, with the constraint that all values must be greater than or equal to zero. All algorithms are implemented using the scikit-learn python library.[39]

While reasonable two-, three-, and four-factor NMF solutions exist, here we focus on the three-factor solution, chosen by use of a cross-validation approach[40−42] (described in detail in section 4 of the Supporting Information). As shown below, the three-factor solution provides a straightforward interpretation of the data, though we note that choosing the two- and four-factor solutions (given in section 6 of the Supporting Information) would lead to similar overall conclusions. The chemical makeup of each factor is described by the fraction of each chemical species associated with it, as shown in Figure 2a. Factor 1 is dominated by the CO signal (including >95% of the total CO) with additional contributions from SO_2, NO_2, and particles in bin 0, suggesting this factor is indicative of combustion-related activity. Factor 2 is associated with a substantial fraction of the bin 0 particles and reactive gases (O_3 annd NO_2), with a smaller contribution from the particles in bins 1 and 2. Factor 3 is associated mostly with the bin 2 particles, with smaller contributions from the other particle bins, and virtually no contributions from gas-phase pollutants. As shown in the 4 day time series (Figure 2b), factor 1 has a distinct diurnal behavior, with a maximum in the early afternoon. The two particle-dominated factors have qualitatively similar diurnal profiles, but their ratios vary considerably throughout the measurement period (see section 5 of the Supporting Information), suggesting they are not simply a single split factor. Throughout the rest of this paper, we will refer to these factors as the CO-dominated factor, particle factor 1, and particle factor 2, respectively.

To explore the potential relationships between these LCS-derived factors and pollution sources, we correlate the results of each factor's time series to the physical and chemical properties of the aerosol, as measured by the research-grade instruments. These metrics include the particle number concentration (N_{tot}) and mass concentration (PM_1) as measured by the SMPS and the aerosol chemical composition (Org, SO_4, NO_3, NH_4, Cl, and BC) as measured by the ACSM and Aethalometer. Figure 3a shows the correlation (as Pearson r^2) of the NMF factors with each of these measurements. The CO-dominated factor correlates strongly (Pearson $r^2 \sim 0.7$) with the total number of particles (N_{tot}) as well as both the organic and black carbon fractions of PM_1, with little to no correlation with the inorganic component of the aerosol. This result is consistent with our interpretation of this factor indicating primary combustion emissions, though the organic component may include secondary organic aerosol (SOA), as well. Notably, this combustion factor includes very little contribution from the PM measurements; this is likely because combustion-derived particles are too small to be detected by the OPC (which has a size cutoff of ∼400 nm). Indeed, the high minimum cutoff diameter represents a major limitation of current low-cost PM sensors. However, as shown here, LCS measurements of gaseous pollutants nonetheless enable the inference of these particles and their sources.

Particle factor 1 correlates most strongly with ammonium and chloride ($r^2 > 0.7$) and less strongly with sulfate and nitrate; it does not correlate at all with the organic or black carbon fractions of PM_1, suggesting this factor is indicative of secondary inorganic species and is more regional in nature than the combustion factor.[21,22] Particle factor 2 also correlates well with the secondary inorganic species with a stronger correlation to sulfate and a weaker correlation to chloride and nitrate than particle factor 1. Both of these factors correlate with PM_1 mass, likely because the secondary inorganic species reside in the upper part of the accumulation mode where a substantial fraction of the total PM_1 mass resides (see section 8 of the Supporting Information).

Figure 3b, which shows the correlation between the LCS NMF factors and the (independently determined) ACSM PMF factors, provides additional support for these assign-

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ments. Specifically, the CO-dominated factor correlates strongly ($r^2 \sim 0.9$) with the primary organic aerosol (POA = HOA/BBOA) factor, which constitutes more than 50% of the organic aerosol (OA) mass during this measurement period.[21] Interestingly, the remaining OA is only slightly captured by our factor analysis; this may be because the SOA present in the lower part of the accumulation mode (100−400 nm) is not visible to our OPC and does not correlate with any of the gas-phase measurements taken by the MPAQS.

Both particle factors correlate well with the ammonium chloride (AC) factor from the ACSM, as it is the factor that captures the bulk of the secondary inorganic species and likely resides in the upper part of the accumulation mode that can be measured by the OPC. The two particle factors also show some difference in their (weak) correlation to the SOA and inorganic factors (ANOOA and ASOOA), which could be related to the changes in aerosol composition due to a combination of local and regional sources throughout the day, though a full understanding of this effect requires further investigation.

Despite the limitations that many LCS present, including their inability to measure ultrafine particles, various types of interference associated with electrochemical sensors, and generally uncertain calibrations, this work demonstrates how multipollutant sensor data can yield interpretable insights into the sources and composition of air pollution. The combination of gas- and particle-phase sensors into a single instrument provides more information than is possible from any single sensor type alone. In this case study, the LCS agree with, and are even complementary to, the research-grade instrumentation commonly used in atmospheric chemistry studies. In particular, multipollutant LCS provide information about particle sources despite being unable to measure most of the particles, by leveraging the clear, strong signal in the gas phase; in this case, elevated CO levels are indicative of combustion emissions, which include primary organic particles that are too small to be detected by low-cost particle sensors. Overall, this work demonstrates the utility of using LCS not only for measuring air quality and pollutant levels but also for understanding sources of pollution and for providing insights into its underlying atmospheric chemistry.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.9b00393.

> Detailed information about the sensor hardware, assessing sensor-to-sensor variability, choosing the optimal number of NMF factors, alternative NMF solutions (two- and four-factor solutions), results for two replicate sensors, estimating error, estimating PM1 loadings from reference instruments, and complete time series (PDF)

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Notes

The authors declare the following competing financial interest(s): D.H.H. is the co-founder of QuantAQ, Inc., which manufacturers air quality sensors and associated software. None of the work completed for this study was done while the entity existed (founding date, June 10, 2019), though the company was founded prior to publication. The remaining authors declare no competing financial interest.

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