Environmental Science & Technology

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# The Effect of a Receding Saline Lake (The Salton Sea) on Airborne Particulate Matter Composition

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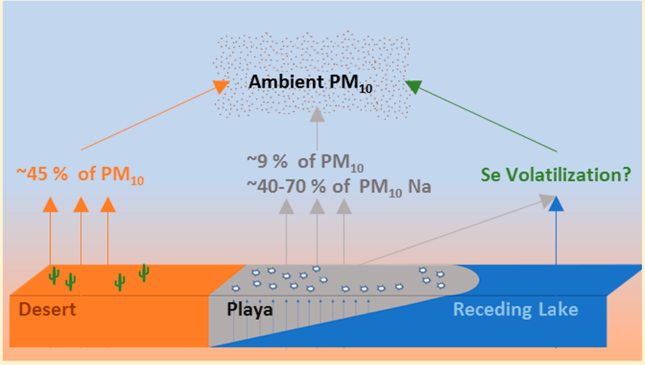
*Supporting Information*

**ABSTRACT:** The composition of ambient particulate matter (PM) and its sources were investigated at the Salton Sea, a shrinking saline lake in California. To investigate the influence of playa exposure on PM composition, PM samples were collected during two seasons and at two sites around the Salton Sea. To characterize source composition, soil samples were collected from local playa and desert surfaces. PM and soil samples were analyzed for 15 elements using mass spectrometry and X-ray diffraction. The contribution of sources to PM mass and composition was investigated using Al-referenced enrichment factors (EFs) and source factors resolved from positive matrix factorization (PMF). Playa soils were found to be significantly enriched in Ca, Na, and Se relative to desert soils. PMF analysis resolved the $PM_{10}$ data with four source factors, identified as Playa-like, Desert-like, Ca-rich, and Se. Playa-like and desert-like sources were estimated to contribute to a daily average of 8.9% and 45% of $PM_{10}$ mass, respectively. Additionally, playa sources were estimated to contribute to 38–68% of $PM_{10}$ Na. $PM_{10}$ Se concentrations showed strong seasonal variations, suggesting a seasonal cycle of Se volatilization and recondensation. These results support the importance of playas as a source of PM mass and a controlling factor of PM composition.



## 1. INTRODUCTION

The Salton Sea is an endorheic, saline lake, located in the southwest of the United States. The Sea has been intermittently filled with water on geological time scales and was last filled through an accidental diversion of the Colorado River in 1905.[1] For the past century, inflows dominated by agricultural runoff and municipal effluent maintained Salton Sea water levels. In 2003, the Quantification Settlement Agreement (QSA) incentivized agricultural water conservation in the region, resulting in a decrease in runoff inputs to the Sea. As part of the QSA, additional water diverted from the Colorado River is directed into the Sea to replace inflow reductions. The diversion of river water is scheduled to end before 2018, and the decrease of inflow is predicted to cause a significant decline in water level between 2018 and 2030.[2] This drop will expose large areas of dry lakebed (playa), potentially creating a vast area of highly emissive surfaces in a county that is already classified as nonattainment for particulate matter with aerodynamic diameters ≤10 μm ($PM_{10}$).[3] Due to drought, large areas of playa are already exposed and are acting as dust sources with an unknown effect on PM mass or composition.

Globally, dust emissions from exposed playas have negatively affected air quality in areas including Owens Lake, California, Sevier Lake, Utah, and the Aral Sea, Uzbekistan and Kazakhstan.[4−6] Playas have a high potential to act as dust sources because playa surfaces are often unvegetated and typically have elevated levels of subsurface capillary action that generate irregular and emissive salt crusts.[7] The dust emissions from playas increase airborne particulate matter (PM) mass and raise health concerns, as increases in PM mass have been linked to cardiovascular disease, respiratory disease, and mortality.[8] In addition to increased mass loadings, the composition of PM emissions could increase particulate toxicity.[9−11] There is concern that metals and pesticides from Salton Sea sediments may be suspended in dust and increase PM toxicity; selenium, cadmium, molybdenum, nickel, and zinc and have all been previously observed at levels of ecological concern within Salton Sea Sediments.[12,13] Additionally, in other playas contaminants have been observed to accumulate after desiccation, potentially intensifying the toxicity of emissions. For example, at Owens Lake and Ash Meadows, accumulation of As has been observed in near-surface sediments and dust emissions.[4,14] PM sourced from playas is thought to be compositionally distinct from PM sourced from other dust sources, but the magnitude of changes to bulk PM composition

**Received:** April 6, 2017
**Revised:** July 10, 2017
**Accepted:** July 12, 2017
**Published:** July 12, 2017

DOI: 10.1021/acs.est.7b01773

has not been investigated at the Salton Sea or in other major playa systems.

Here we test the hypothesis that emissions from playas change $PM_{10}$ composition near the Salton Sea, potentially influencing toxicity, dust microphysical processes, and nutrient deposition. We assess the composition of playa soils, desert soils, and $PM_{10}$ collected at the Salton Sea during August 2015 and February 2016. Major sources of PM are identified using enrichment factors and positive matrix factorization (PMF), and the mass contribution of desert dust and playa dust sources are estimated.

## 2. METHODOLOGY AND APPROACH

**2.1. Sampling.** The locations of all aerosol and soil sampling sites are shown in Figure S1 of the Supporting Information (SI). Aerosol samples were collected at Salton City (33.27275°N, −115.90062°W), CA and Bombay Beach (33.35264°N, −115.73419°W), CA, two population centers along the coast of the Salton Sea. These sites were chosen due to their proximity to the Salton Sea and the presence of Imperial Irrigation District (IID) air quality monitoring stations, which enabled easy sample collection and access to bulk $PM_{10}$, wind speed, wind direction, relative humidity (RH), and temperature data. Meteorological data were obtained from the California Environmental Protection Agency's, Air Resources Board, Air Quality and Meteorology Information System.[15] Aerosol samples were collected at Salton City (SC) during Aug. 22−27, 2015 (SCS) and Feb. 12−19, 2016 (SCW) and at Bombay Beach (BB) during August 27−31, 2015 (BBS). During both sampling periods, the Salton Sea water level averaged ∼71.4 m below sea level, corresponding to about 15 000 acres of exposed playa relative to year 2000 levels.[2,16] Aerosol samples were collected in two sets to capture the influence of different daily wind patterns: a day set between 08:00 LT (local time) and 18:00 LT and a night set between 19:00 LT and 07:00 LT. All samples were collected by a Micro-Orifice Uniform Deposition Impactor (MOUDI) sampler, which separates and collects aerosols by size into 11 stages, between 0.056 and 18 μm, onto 47 mm, 2 μm pore diameter, Pall Zefluor PTFE filters. During sampling, three summer and two winter sets of field blanks were also collected. Pre- and postsampling, filters were kept in acid-washed Petri dishes and stored individually in antistatic bags. Data from the night of August 24th and the day of Aug. 25th were not included due to water condensation within the MOUDI.

A total of 25 playa and 88 desert samples were collected from a wide area around the Salton Sea. Playa sites are characterized by a lack of vegetation and were recently submerged underneath the Salton Sea, whereas Desert sites are located farther from the sea. Soils were collected by sampling the top 1 cm layer of the surface soil- the section that would most likely undergo suspension. Soils were stored in quart Ziploc bags and left open to air-dry at 22 °C.

**2.3. Filter Digestion and Elemental Analysis.** Soil (7 playa and 10 desert) and aerosol samples were acid digested to determine total metal and metalloid concentrations for enrichment factor calculations and as inputs for the PMF model. For soil samples, a small mass of soil (∼500 μg) was digested. To increase the signal-to-noise ratio within aerosol samples, MOUDI filters in the following size ranges were combined for analysis: 0.056−0.18 μm, 0.18−0.56 μm, 1.0−3.2 μm, 3.2−10 μm. Filters collected in the size range of 0.56−1.0 μm were analyzed individually. Filters with particles greater than 10 μm were excluded due to high field blank values. A two-step digestion was performed using $HNO_3$ + HF and aqua regia (i.e., $HNO_3$ + HCl). All acids were sourced from Fisher Chemical and were trace metal grade or better. For each digest, elemental concentrations of the following elements were determined by inductively coupled plasma mass spectroscopy (ICP-MS) (Agilent 7900): Ca, Na, As, Al, Cr, Fe, Mn, Ni, V, Ba, Cd, Co, Se, Ti, K. All analysis was performed using the summed concentrations of extracts from each period to get $PM_{10}$ elemental concentrations. A more detailed description of the digestion procedure and quantification steps is included in the SI.

**2.4. X-ray Fluorescence Spectroscopy.** To confirm representativeness of the smaller sample set analyzed by ICP-MS, a total of 113 soil samples (25 Playa and 88 Desert) were analyzed using energy dispersive X-ray fluorescence (ED-XRF) spectroscopy (SPECTRO XEPOS) to determine total solid phase Na, Al, K, Ca, Ti, V, Cr, Fe, As, Co, and Se concentrations. ED-XRF is commonly used as a high-throughput method to measure total elemental composition in solid phase samples and is useful for investigating overall trends in larger sample sets. ED-XRF was used here as a high throughput method to provide a more representative characterization of both playa and desert samples and to increase confidence in any differences observed between the two soil classifications. A more detailed description of the quantification steps is included in the SI.

**2.5. Enrichment Factors.** Enrichment factors (EF) have been used widely in atmospheric and soil literature to track normalized changes in composition that are driven by anthropogenic or geologic forces.[17,18,5] Enrichment factors are often used to track anthropogenic contamination within PM and soils, but can also be used to monitor natural enrichment and fractionation processes. Mineralogical and chemical transformations can also lead to variations in EFs reflective of specific environmental conditions; thus, interpretation of EF results must be made within the context of the environmental conditions where samples were collected.[19] For this study, Al is used as the normalization element because it is relatively immobile in soils and has been shown to be conserved upon emission. EF in soils and aerosol samples were calculated via

$$\mathrm{EF} = \frac{(M_{\mathrm{measured}}/Al_{\mathrm{measured}})}{(M_{\mathrm{UCC}}/\mathrm{Al}_{\mathrm{UCC}})} \qquad (1)$$

where $M_{\mathrm{measured}}$ is the ICP-MS measured mass concentration of an element within a sample (ppm), $\mathrm{Al}_{\mathrm{measured}}$ is the ICP-MS measured mass concentration of Al within a sample (ppm), and $\mathrm{M}_{\mathrm{UCC}}$ and $\mathrm{Al}_{\mathrm{UCC}}$ are the analogous values for the Upper Continental Crust composition (UCC), as reported by Wedepohl.[20] Samples with enrichment factors of 1 have the same elemental ratios as UCC; values greater than or less than 1 indicate differences from the UCC.

**2.6. Positive Matrix Factorization (PMF).** EPA's Positive matrix factorization (PMF 5.0) is a statistical method that uses factor analysis to determine the contribution of different sources to the measured elements at a receptor site. Unlike principle component analysis, PMF effectively only allows positive values, allowing small negative factor contributions (>-0.2) only to increase the goodness of the model fit. This allows PMF to be applied to environmental data, where negative contributions are not physically meaningful. For PMF, both data and uncertainty matrices are input into the model.

DOI: 10.1021/acs.est.7b01773
*Environ. Sci. Technol.* XXXX, XXX, XXX−XXX

The inclusion of an uncertainty matrix allows the PMF model to provide weights to samples by their signal-to-noise (S/N) ratio and reduce weighting of highly uncertain inputs. More detail about EPA PMF 5.0 can be found in Paatero and Tapper, Paatero, and Brown et al.[21−23] An extended description of the PMF parameters used for this study is included in the SI.

## 3. RESULTS AND DISCUSSION

**3.1. Meteorological Data.** Meteorological data provide an external context for PM measurements, as meteorological factors directly influence playa and desert emissions. For example, humidity and temperature changes can shift hydration states of playa minerals and influence their emissivity.[24] Additionally, wind direction and speed determine strength and likelihood of detection of emissions from specific sources.

Relative humidity (RH) at the sampling sites showed common diurnal variations, with a maximum RH observed in the early morning and a minimum RH in the afternoon (SI Figure S2a and S3). However, the magnitude and range of RH oscillation varied between sampling periods. During Bombay Beach Summer (BBS), Salton City Summer (SCS), and Salton City Winter (SCW) the average daily RH oscillation between 06:00 LT and 15:00 LT was 28.9%, 18.0%, and 10.7%, respectively. The magnitude of RH oscillations is important as larger oscillations lead to larger hydration/dehydration shifts within the evaporite minerals of playa surfaces, weakening salt crusts and increasing emissivity. Given this, playa emissions could be higher during BBS and SCS relative to SCW.

For wind direction analysis, directions were divided into 8 octants (North, Northeast, East, etc.) and probability of wind being sourced from each octant was calculated (Figure 1). Wind direction displayed a clear diurnal trend at all sites (SI Figure S4−S6). Additional discussion of these trends can be found in the SI. It should be noted that the predominant wind directions during day BBS, day SCS, and night BBS were consistent with air masses passing over the Salton Sea and/or playas if a short-term linear path is assumed. If playa emissions were occurring, playa chemical signatures should be observable within samples collected during BBS and SCS. Conversely, the predominant wind directions during SCW would not directly cross the Salton Sea and playa, so playa emissions are not expected to be observed frequently during this period.

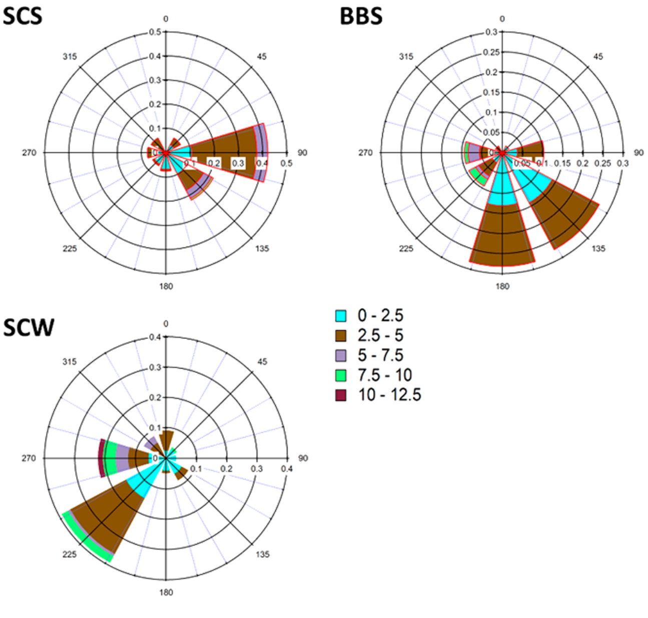


**Figure 1.** Wind roses for sampling periods Salton Sea Summer (SCS), Salton Sea Winter (SCW) and Bombay Beach Summer (BBS). Axis values represent the probability of wind being sourced from an octant and colors represent wind speed in m $s^{-1}$.

Median, hourly averaged, wind speed was 2.55 m $s^{-1}$, 2.62 m $s^{-1}$, and 3.2 m $s^{-1}$ for SCS, BBS, and SCW, respectively. SCW wind speed populations (SI Figure S2c) were skewed toward higher values due to short-term, high wind speed events occurring on 2/14/16 and 2/18/16. No significant wind speed differences were observed between day and night periods for SCW and BBS, but a significant difference was observed for SCS. For SCS, median day and night wind speeds were 3.2 m $s^{-1}$ and 1.3 m $s^{-1}$, respectively.

**3.2. Soils Characteristics.** ED-XRF and ICP-MS elemental analysis revealed consistent and unique characteristics of playa and desert soils (SI Table S1 and S2). For both techniques, playa soils were observed to have significantly ($p \leq 0.05$) larger elemental abundances of Na, Ca, and Se and desert soils were observed to have significantly larger abundances of Ti and Mn. The larger ED-XRF data set also revealed significantly greater abundances of Al, Fe, K, V, and Cr within desert soils. Notably, arsenic was not significantly different between playa and desert soils using either techniques, and Cd was observed at near or below detection limits of both techniques. Given these trends, increased emissions of PM from playas are expected to increase abundances of Na, Ca, and Se in dust particles relative to typical desert soil emissions. Because of the observed consistencies in ED-XRF and ICP-MS analysis of the samples, and to allow intercomparison with the PM data, only ICP-MS data were used for EF calculations.

**3.3. $PM_{10}$ Data.** Median, hourly $PM_{10}$ mass concentrations from tapered element oscillating microbalance (TEOM) (Thermo-scientific, SER 1400) measurements during BBS, SCS, and SCW were respectively 28.7 $\mu$g $m^{-3}$, 23.3 $\mu$g $m^{-3}$, and 15.6 $\mu$g $m^{-3}$ (SI Figure S7). Summer (BBS and SCS) $PM_{10}$ concentrations were significantly ($p \leq 0.05$) higher than winter (SCW). Baseline concentrations were lower during winter months, but during SCW, two high mass events were observed on 2/18/16 and 2/14/16, skewing the distribution toward higher mass concentrations, likely due to higher wind speeds observed on these days. Significant diurnal variability in $PM_{10}$ mass concentration was only observed during SCS, with median loadings of 25.7 $\mu$g $m^{-3}$ and 20.4 $\mu$g $m^{-3}$ (SI Figure S8) for night and day, respectively.

*3.3.1. Elemental Mass Concentrations.* $PM_{10}$ median mass concentrations of elements measured via ICP-MS are included in SI Table S3. Mass concentrations of major elements (Al, Fe, Na, and Ca) were found to be within the range of previously observed values in rural areas of the American southwest.[25,26] Daytime vs nighttime trends at the two sampling sites are not explicitly discussed because they were not qualitatively different.

Although trace elements, for example, Cd, Se, Cr, As, Mn, and Ni, do not significantly increase the mass of PM, their concentrations are of interest due to their potential to increase toxicity. Median mass concentrations of each of these metals were below California EPA reference exposure levels (RELs) for chronic toxicity, by factors of 77, 14,000, 18, 250, 7, and 2 respectively (SI Table S3).[27] Nickel was the only element to breach its REL during sampling, during 3 of the 25 sampling periods. This suggests that PM mass increases may be the first

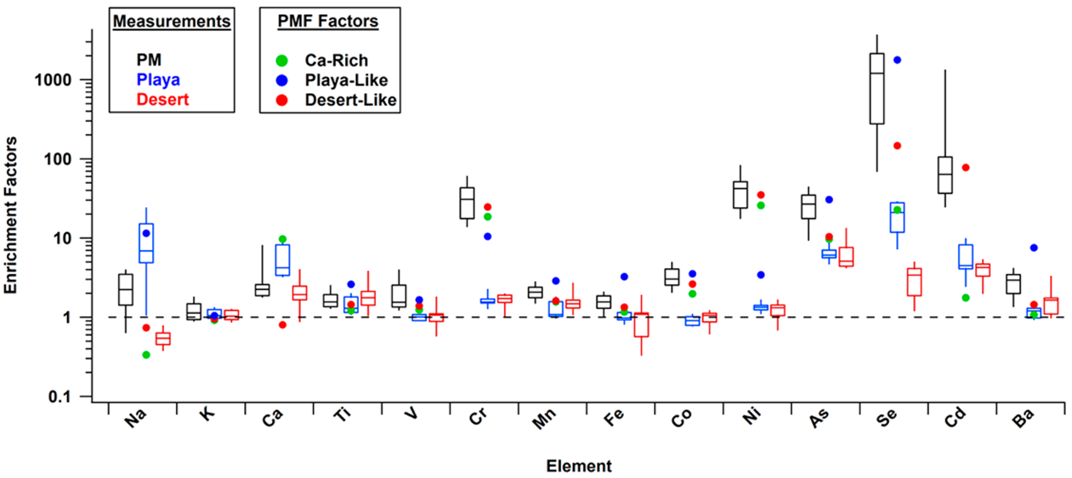


**Figure 2.** Enrichment factor distributions for each element from measurements of $PM_{10}$ (black), playa soils (blue), and desert soils (red). Box and whiskers highlight 10th, 25th, 50th, 75th, and 90th percentiles. Respective EFs of the PMF modeled Ca-rich (green dots), playa-like (blue dots), and desert-like (red dots) factors superimposed. EF of the Se factor was not graphed because low Al attributions create nonmeaningful EFs.

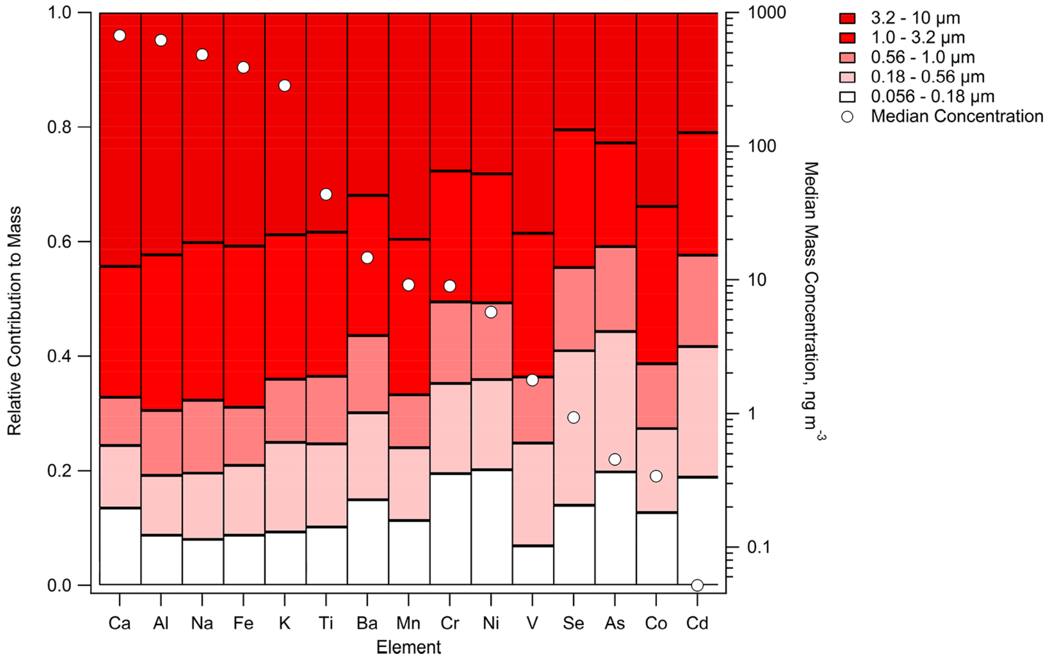


**Figure 3.** Relative mass contribution of different size classes of aerosols to individual elements (left axis) and median mass concentrations of individual elements (right axis).

health concern driven by playa emissions until individual elemental limits are surpassed. The disconnect between sediment contamination and atmospheric PM toxicity likely arises from the different pathways driving PM human toxicity and sediment-driven aquatic toxicity. The metals previously observed at levels of ecological concern in sediments were classified as such, in part, because of their ability to bioaccumulate within aquatic systems.[13,12,28] Conversely, PM toxicity is driven by size and composition of PM,[29] not compounding within the food chain. The crusts that form on playas are not simply dried sediments, but instead are new structures with high concentrations of evaporite minerals.[14,7] Therefore, the presence of contaminants within sediments does not always equate to toxic playas or atmospheric emissions.

Only Na and Se displayed significant differences ($p \leq 0.05$) between summer (SCS+BBS) and winter (SCW) sampling (SI Table S3). Average Na concentrations were 850 ± 670 ng $m^{-3}$ and 370 ± 159 ng $m^{-3}$ during the summer and winter, respectively. $PM_{10}$ Na and its seasonality is discussed further in the discussion of EFs (Section 3.3.2). Average Se concentrations were 2.1 ± 2.7 ng $m^{-3}$ in summer and 0.3 ± 0 0.4 ng $m^{-3}$ in winter. Possible drivers of Se seasonality are discussed in section 3.4.

### 3.3.2. Enrichment Factors (EFs).

EF distributions for playa soil, desert soil, and $PM_{10}$ are shown in Figure 2 and values are provided in SI Table S4. Playa soils were significantly ($p \leq 0.05$) more enriched in Na, Ca, Se than desert soils. Increased concentrations of these elements have previously been observed within Salton Sea sediments and waters;[30,31,13] these elements were likely deposited during the playa formation.

$PM_{10}$ was significantly more enriched than both soil types in Se, V, Cr, Ni, As, Ba, and Fe. Enrichment of the minor elements

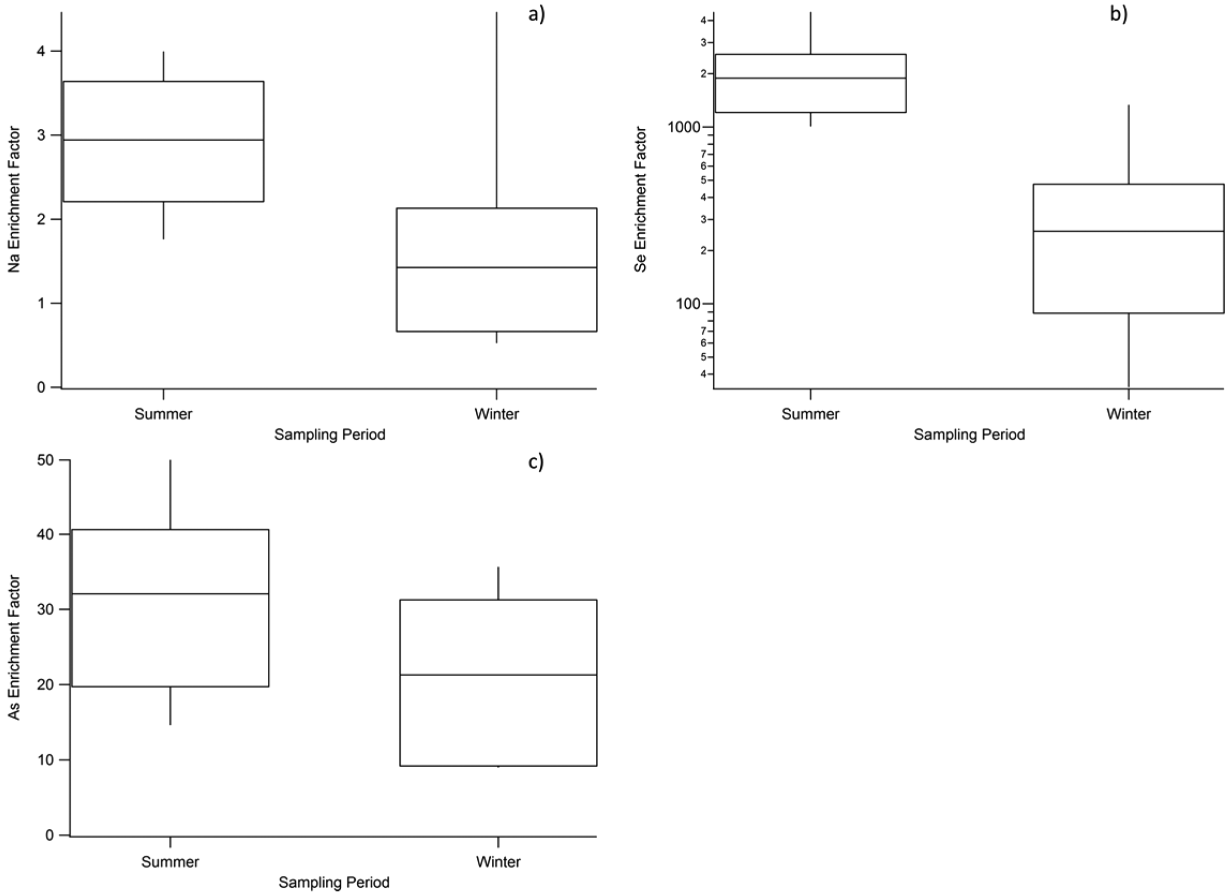


**Figure 4.** $PM_{10}$ EF distributions of (a) sodium, (b) selenium, and (c) arsenic- elements with significant seasonal differences in EF distribution. Box and whiskers highlight 10th, 25th, 50th, 75th, and 90th percentiles.

has been previously observed during simulated suspension of soil in the laboratory.[5,32] A soil suspension enrichment may have contributed to the observed enrichments in $PM_{10}$. As shown in Figure 3, minor and trace elements in $PM_{10}$ were concentrated on smaller particles while crustal elements (Fe, Al, Ca, Ti) were concentrated on larger particles. Anthropogenic sources including vehicles (Fe, Pb, V), incinerators (Ni, As), biomass burning (K, Zn) and coal combustion (Se, Pb) are also possible contributors to metal enrichment.[33,13,34,35] Iron was the only major element significantly enriched in $PM_{10}$ relative to both soil types. Traffic emissions present one possible source of Fe enrichment, as Fe has been observed in brake pad and vehicular emissions.[36,37] A combination of suspension enrichment and anthropogenic contributions are reflected in the enrichments of $PM_{10}$.

$PM_{10}$ was significantly enriched in Na relative to desert soils, but not playa soils. $PM_{10}$ Na concentration is of interest because Na containing minerals including, but not limited to, halite (NaCl) have been observed in high concentrations within playas.[24,38,39] This study also observed Salton Sea playas with significantly ($p \leq 0.05$) higher Na concentrations than local desert soil (Section 3.2). Emissions from playa areas are expected to have relatively high Na concentrations, so Na can be utilized as a tracer for these emissions. $PM_{10}$ Na enrichment may arise from mixing of two sources, that is, playa and desert soil. This enrichment suggests that playa emissions are contributing significantly to the Na within $PM_{10}$. The percentage of sodium attributed to playa emissions is discussed in section 3.5.

Summer (SCS+BBS) $PM_{10}$ EFs were significantly greater than winter (SCW) for Se, Na, and As (Figure 4). The lack of a seasonal pattern in the EFs of major elements other than Na suggest that separate factors control the emissions of Na compared to other major crustal elements. This would be expected if the Na was sourced from playa, as playa emissions are thought to be influenced by additional meteorological factors such as RH,[24] while dust emissions from nonplaya sources are not.[40] Higher Na EFs during summer samplings are consistent with wind direction and RH observations, as wind directions during both SCS and BBS were predominantly from the direction of playas and larger diurnal RH oscillations were observed during these periods. Oscillations in RH are thought to affect the hydration state of playas, increasing irregularity in mineral structure and potentially increasing emissivity. Na EFs were weakly correlated with EFs of Se ($r = 0.5$, $p = 0.01$) and As ($r = 0.4$, $p = 0.04$), indicating a large proportion of Se and As were likely emitted from sources separate than those of Na. Selenium displayed the largest enrichment among the trace elements in $PM_{10}$ and the strongest seasonal dependence: median summer and winter Se $PM_{10}$ EFs were 1890 and 257, respectively.

**3.4. Positive Matrix Factorization (PMF).** The four source factors produced by PMF, using precedents set previously in published source studies and knowledge of potential sources in the area, were identified as playa-like, desert-like, Se, and Ca-rich. Profiles of these factors are shown in Figure 5. Notably, elements normally associated with anthropogenic emissions (such as V, As, Ni, Cr, and Ba),

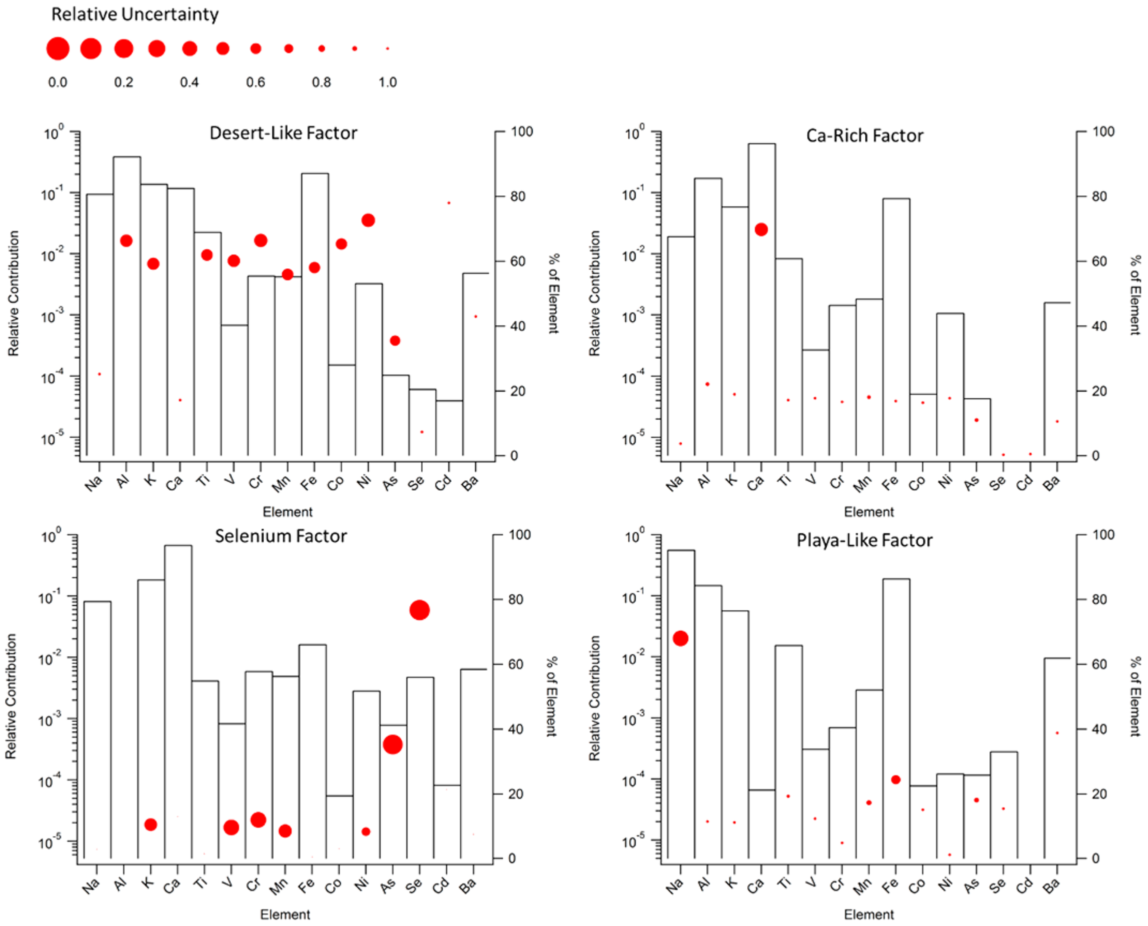


**Figure 5.** Positive matrix factorization (PMF) factor profiles, with the relative contribution of elements on the left axis, represented by bars, and the percentage of each element accounted for by a factor on the right axis, represented by red dots. The size of the red dots is inversely proportional to the relative uncertainty, and represents the confidence in the percentage mass of each element accounted for by each factor, with larger dots representing more confident attributions. The uncertainty is calculated by the displacement method.

which were observed to be enriched in PM relative to playa and desert soils, were not isolated in an independent factor. This is likely due to the low concentrations and relatively high uncertainties associated with these elements.

The playa-like factor was characterized by a prominence of Na and the presence of dust tracers such as Fe, K, and Ti. The Na and K EFs of the playa factor were also within the distribution of EFs observed for playa soils, although EFs of other elements were higher than those observed in playa soils. These high enrichments likely arise from the high uncertainty in the attribution of elements other than Na to this factor. The playa-like factor is similar to sea spray factors, typically dominated by high concentrations of Na and/or Cl, observed in other PMF studies.[41,42] Here, the playa-like factor is not believed to be sea spray as conditions for white-cap formation, that is, wind speeds greater than 4 $\text{ms}^{-1}$, were not observed in high frequency during any sampling period, except for SCW.[43] On average, the playa-like factor contributed the least to samples from the SCW period when higher wind speeds were occasionally observed, suggesting a source different than sea spray. Furthermore, if sea spray were an important source, $PM_{10}$ EFs of Na would be expected to be higher than those observed in the playa on at least some of the sampling days, but this was not observed. Similar to trends observed within the $PM_{10}$ EF of Na, the contribution of the playa-like factor was higher on sampling days with the wind blowing over playa surfaces.

The desert-like factor was identified by the prominence of Al, Fe, K, and Ti, all of which are major elements in the Earth's crust and have been observed to be prominent in regional deserts' soils (Section 3.1),[20] and have been associated with crustal emissions in previous PMF studies.[44−46] Observation of a desert-like factor is expected because dust sourced from desert surfaces is thought to be a large component of PM mass in the rural southwest.[24,25] The desert-like factor's EFs of major elements (Na, K, Ti, Mn, Fe) were within the distributions of EFs for sampled desert soils (Figure 2), further supporting the conclusion that this factor represents the contribution of local soils. Notably, the desert-like factor was much more enriched in Co, Ni, Se, Cd, and Cr than the desert sample set (Figure 2); this likely occurs because no separate anthropogenic source was resolved. These elements are commonly associated with traffic, industry, and incineration and should not be interpreted as being sourced from natural dust emissions.

The Ca-rich factor is similar to Ca factors found in previous source apportionment studies and often attributed to construction or resuspension of road dust.[47,48] The sampling sites were close to population centers and an off-road vehicle park, so it is possible that anthropogenic activity also contributed to this factor. Additionally, other studies have attributed Ca-rich factors to secondary dust sources, such as limestone or gypsum deposits.[49,50] The Salton Sea and surrounding areas are found in the basin of the ancient lake Cahuilla, and the lacustrine deposits could contribute to a secondary dust source and this factor. Gypsum and other Ca-containing minerals are also present within Salton Sea playas,[24] but on days when the Ca-rich factor was most prominent, wind direction was not predominantly from the direction of the Salton Sea. These observations suggest that Salton Sea playas are not the major source of the Ca-rich factor and that it is likely a combination of anthropogenic and natural Ca sources.

The Se factor was identified by the prominence of Se; 77% of all sampled Se is attributed to this factor. Given that coal combustion is the major anthropogenic source of Se and the lack of coal usage in this region, the Se factor was likely not anthropogenic in nature. One potential Se source is Se volatilization and condensation onto PM.[51] High levels of Se volatilization would explain both the source independence and seasonal variability observed in Se PM mass concentrations, EFs, and the Se factor. Se volatilization has been previously observed in submerged sediments and capillary fringe soils, like those found in the subsurface of the playas.[52−55] Previous observations of Se volatilization report strong seasonal variability, with the highest rates of volatilization occurring during summer months. Summer median Se EFs were 7 times greater than winter, suggesting that Se volatilization from the sediments/soil and condensation onto PM is an important process at the Salton Sea. Previous investigations of Se volatilization at the Salton Sea by Schroeder et al. (2002) reported gradients of volatile Se species occurring within the water column, providing evidence that volatilization within sediments was occurring.[31] Despite this, Se volatilization from sediments was not previously thought to play a large role in the Salton Sea Se budget and mass balanced was achieved accounting for Se only within the aqueous phase, sediments, and biomass. If Salton Sea playa zones have a greater rate of Se volatilization than Salton Sea sediments, volatilization may become increasingly important during the exposure of playas. These data reveal that Se dynamics at Salton Sea need to be further investigated, as volatilization pathways may be stronger than previously estimated. These findings may have implications for the Salton Sea restoration plans.

### 3.5. Two-Source Mixing Model and Na Attribution.

If playa and desert sources are the dominant sources of Na and Al in PM at the Salton Sea, it is possible to construct a simple two source mixing model (TSM). Using the median playa and desert soil EFs as end points, the contribution of each respective source to Na can be estimated via:

$$\mathrm{EF}_{\mathrm{PM}} = \mathrm{EF}_{\mathrm{Playa}}(X) + \mathrm{EF}_{\mathrm{Desert}}(1 - X) \tag{2}$$

where $\mathrm{EF}_{\mathrm{PM}}$ is the observed Na EF for a sample within $\mathrm{PM}_{10}$, $\mathrm{EF}_{\mathrm{playa}}$ is the median Na EF of playa soils (6.9, standard deviation (SD) = 7.8) and $\mathrm{EF}_{\mathrm{desert}}$ is the median Na EF of desert soils (0.54, SD = 0.13). $X$ is the percentage of $\mathrm{PM}_{10}$ Na attributed to playa sources during a given sampling period.

TSM-based playa Na concentrations correlate very well ($r$ = 0.96, $p = 1.4 \times 10^{-14}$) with the playa-like Na concentrations from PMF. A high correlation between playa Na concentrations estimated by TSM and PMF confirm that a two-source system of soils and playa, and not sea salt, indeed controls the $\mathrm{PM}_{10}$ Na concentrations in the basin. Despite high correlations, TSM estimates lower sodium emissions from Playa (38% of $\mathrm{PM}_{10}$ Na) than PMF (~68% of $\mathrm{PM}_{10}$ Na). It is possible that TSM underestimates the playa contribution to Na as $\mathrm{EF}_{\mathrm{playa}}$ is highly variable (SD = 7.8). To match the TSM and PMF-based estimates of playa contributions to Na, $\mathrm{EF}_{\mathrm{playa}}$ in eq 2 needs to be lowered to 4.2, which is still within the observed range of Na $\mathrm{EF}_{\mathrm{playa.}}$ The large variation in playa composition likely leads to a similarly large distribution of playa emission potentials and compositions. Therefore, the effective Na $\mathrm{EF}_{\mathrm{playa}}$ of 4.2 may in fact be a more representative value of the Na EF of emissive playas.

### 3.6. Crustal and Playa Contributions to $\mathrm{PM}_{10}$.

To quantify the burden of $\mathrm{PM}_{10}$ emission sources at Salton Sea, the mass contributions of major sources need to be calculated. Elemental masses do not represent the complete mass represented by a factor, especially for mineral-based factors like the playa-like, desert-like, and Ca-rich factors because the digestion and analysis method could not measure bulk Si-, O-, N-, or C-based species. Despite this, PM mass associated with the Desert-like factor can be estimated using the elemental concentration of Al, Ca, Fe, and Ti in dusts and the formulation from the Interagency Monitoring of Protected Visual Environments (IMPROVE) program and modified by Clements et al.[56−58,25]

$$\text{crustal mass} = 10.92(\mathrm{Al}) + 1.63(Ca) + 2.42(\mathrm{Fe}) + 1.94(Ti) \tag{3}$$

where crustal mass is the $\mathrm{PM}_{10}$ mass concentration from crustal sources and Al, Ca, Fe, and Ti are the elemental mass concentrations of each element observed in $\mathrm{PM}_{10}$. Using this method, and averaging over the sampling periods, the average crustal contribution was 45.2 ± 26.4% (13 ± 10 $\mu$g m$^{-3}$) of the total $\mathrm{PM}_{10}$ mass, as measured by the IID, indicating that on most days crustal sources played a major role in the observed $\mathrm{PM}_{10}$ mass loadings at these sites.

An integral goal of this study was to determine the $\mathrm{PM}_{10}$ mass contribution of playa sources around Salton Sea. No analogous equation exists to estimate playa contributions, in part because playa composition is variable between sites. Here, an equation was developed using our PMF results and the playa mineral frequencies described by Buck et al. (2011).[24] Assuming this subset is representative of emissive playa areas around the Salton Sea, the ratio of Na to total mineral mass of a typical playa can be calculated. Next, using the PMF estimated Na concentration of the playa-like factor, the $\mathrm{PM}_{10}$ mass contribution of playa sources can be estimated by

$$\text{Playa mass} = k \times \mathrm{Na}_{\mathrm{Play}a} \tag{4}$$

where Playa mass is the $\mathrm{PM}_{10}$ mass concentration sourced from playas on a given day, $k$ is 5.22 and represents the ratio of total playa mass to Na mass as observed by Buck et al. (2011), and $\mathrm{Na}_{\mathrm{Playa}}$ is the mass concentration of Na that was attributed to playa sources by PMF on the day of interest. Using this estimation, playa sources contributed at an average of 8.9 ± 5.6%, (2.2 ± 1.6 $\mu$g m$^{-1}$) to $\mathrm{PM}_{10}$ mass over all sampling periods. Although highly variable, average summertime contribution of playa sources to $\mathrm{PM}_{10}$ mass at both sites was higher than in winter (11.9 ± 11.4% vs 5.13 ± 5.8%). The contribution of playa to PM at these sites is not negligible, but measurable at current levels of playa exposure. These contributions will likely increase with increasing exposures of the shoreline.

High dust emission events with prevailing winds crossing over the exposed playas were not sampled during this campaign. Since PM dust emissions caused by wind erosion

DOI: 10.1021/acs.est.7b01773

are controlled in part by wind speed and the presence of saltating particles, playa emission rates are expected to increase under higher wind conditions. Therefore, the observations here may only represent the lower bounds of current playa contributions to $PM_{10}$. Future PM source apportionment studies need to resolve the contributions of playa areas during high mass events, as these contributions will likely be increasingly more important as the Salton Sea recedes and shoreline exposures grow in extent.

Notably, our results do not support that concept that playa emissions or PM in this region, are exceptionally toxic due to high levels of toxic metals (Section 3.3.1). Despite this, both TSM and PMF results provide evidence for a relatively strong impact of playa on PM Na concentrations and $PM_{10}$ mass loadings. Such emissions could influence downwind biogeochemical and atmospheric processes and affect ecology and meteorology in downwind regions. For example, Na deposited in soils can influence biomass decomposition and perturb plant nutrient balance.[59,60] While suspended as PM, Na can also impact cloud formation.[61] With recent playa exposures in other regions, for example, the Great Salt Lake in southwest U.S.,[62] Ebinur Lake in northern China,[63] and Lake Urmia in Iran,[64] methodologies applied here can be used to determine PM compositional shifts and estimate the impact of playa exposures on PM concentration due to changes in lake size.

## ■ ASSOCIATED CONTENT

### Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b01773.

> Detailed methodology concerning digestions, ED-XRF and PMF; descriptions of temperature and wind conditions; Tables S1–S4: summary tables of elemental measurements and EFs; Figures S1–S9: map of sampling locations; multiple graphics presenting meteorological conditions (PDF)

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### Notes

The authors declare no competing financial interest.

## ■ ACKNOWLEDGMENTS

We acknowledge the assistance of the Imperial Irrigation District staff who were crucial for logistical support and implementation of this project, as well as Andrew Robinson and Steve Bates (UCR, Department of Earth Sciences) who provided technical expertise regarding sample digestion techniques and ICP-MS analysis. This material is based upon work supported by UCR Regents' Faculty Development Award, USDA National Institute of Food and Agriculture, Hatch project Accession No. 233133, UCR Provost Research Fellowship, and the U.S. Geological Survey under Grant/Cooperative Agreement Nos. G11AP20121 and G16AP00041. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the opinions or policies of the U.S. Geological Survey. Mention of trade names or commercial products does not constitute their endorsement by the U.S. Geological Survey.

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