Changes in speciated PM$_{2.5}$ concentrations in Fresno, California, due to NO$_x$ reductions and variations in diurnal emission profiles by day of week

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The San Joaquin Valley in California suffers from poor air quality due to a combination of local emissions and weak ventilation. Over the course of decades, there has been a concerted effort to control emissions from vehicles as well as from residential wood burning. A multiple linear regression model was used to evaluate the trends in air pollution over multiple time scales: by year, by season, by day of the week and by time of day. The model was applied to 18 years of measurements in Fresno including hourly mole fractions of NO$_x$ and concentrations of PM$_{2.5}$; and daily measurements of speciated components of PM$_{2.5}$. The analysis shows that there have been reductions in NO$_x$, elemental carbon and ammonium nitrate of 4 to 6%/year. On weekends, NO$_x$ mole fractions are reduced by 15 to 30% due to fewer vehicle miles traveled and a smaller fraction of diesel traffic. These weekend reductions in NO$_x$ have not been accompanied by weekend reductions in PM$_{2.5}$ however. In particular, elemental and organic carbon concentrations are higher on winter weekends. Analysis of diurnal profiles suggests that this is because of increased PM$_{2.5}$ on Saturday and holiday evenings which are likely due to residential wood combustion. Furthermore, while organic carbon concentrations have decreased in the winter months, they have been variable but without a net decline in the summer, most likely as a result of forest fires offsetting other improvements in air quality. Fog was found to greatly enhance ammonium nitrate formation and was therefore associated with higher PM$_{2.5}$ in the winter months. Overall the analysis shows that air quality controls have been effective at reducing NO$_x$ all year and PM$_{2.5}$ in the winter, that continued reductions in emissions will further reduce pollutant concentrations, but that winter residential wood combustion and summer forest fires could offset some of the gains obtained.

Keywords: Speciated PM$_{2.5}$; NO$_x$; Diurnal profiles; Mobile sources; Residential wood combustion; Multiple linear regression
The San Joaquin Valley has been extensively studied using long-term monitoring stations, specialized field campaigns and modeling studies (Hu et al., 2014; Reynolds et al., 2012; Chow et al., 2006; Watson and Chow, 2000). During the winter months, long residence times lead to high concentrations of primary pollutants, especially PM$_{2.5}$. During the summer months, high solar radiation leads to ozone exceedances. As these air quality issues are very distinct, they have usually been studied separately.

During the winter, the California Regional Particulate Air Quality Study of 1999 to 2001 found that high PM$_{2.5}$ concentrations consisted of both organic matter and ammonium nitrate particles. The organic aerosols came primarily from local biomass burning such as residential wood burning. The nitrate particles were formed in region wide plumes from the combination of NO$_x$ and ammonia emissions (Chow et al., 2006; Turkiewicz et al., 2006; Hasheminassab et al., 2014).

DISCOVER-AQ took place in the winter of 2013. Measurements of aerosol optical properties confirmed the importance of local residential heating to nighttime PM$_{2.5}$ (Zhang et al., 2016). The nighttime residual layer was also found to be important in the formation of nitrate aerosols which impacted the surface during the morning (Prabhakar et al., 2017) as a result of entrainment mixing (Trousdell et al., 2016).

Modeling studies suggest that the formation of ammonium nitrate was limited by the availability of nitric acid (Kelly et al., 2018) which corroborates the suggestion that controls on nitrogen oxides would be an effective policy for reducing PM$_{2.5}$ (Chen et al., 2014). More recent measurements in the winter of 2015 found that primary biomass burning contributed 27% of organic aerosol mass, and nitrate-related oxidized aerosols contributed 47% on low fog days (Chen et al., 2018). The particle sizes were also found to vary depending on conditions, with wood burning particles increasing in size from the evening into the night (Betha et al., 2018).

Emissions of volatile organic compounds have been reduced from the transportation sector to such an extent that the use of chemical products now makes up a significant fraction of emissions (McDonald et al., 2018). As a result, emissions of volatile organic compounds are thought to be underestimated in the San Joaquin Valley. Modeling studies suggest that updating the emissions would lead to increased reactivity in the winter atmosphere leading to increased formation of ammonium nitrate (Zhu et al., 2019). Measurements and modeling for the Salt Lake Valley suggest that reducing volatile organic compounds may therefore be more effective at reducing ammonium nitrate concentrations than reducing NO$_x$ mole fractions (Womack et al., 2019).

During the summer, the main air quality concern is the occurrence of high ozone mole fractions. Long term reductions in nitrogen oxide emissions have led to reduced ozone exceedances (Balashov et al., 2017). Decreases in nitrogen oxides are also expected to reduce ammonium nitrate concentrations and shift the source of nitrate aerosols from nighttime to daytime chemistry (Pusede et al., 2016). Nevertheless, the understanding of oxidation in the San Joaquin Valley plume was found to have basic uncertainties related to the reactions of nitrogen oxides and hydrogen oxides (Brune et al., 2016). As transportation NO$_x$ reductions have decreased, the relative importance of emissions from other sectors has increased. This is particularly true for NO$_x$ emissions from agriculture which was estimated to increase the NO$_x$ budget in the San Joaquin Valley significantly (Almaraz et al., 2018; Trousdell et al., 2019).

The impact of smoke from wildland fires is another air quality concern during the summer. Although these do not at present cause air quality exceedances most years, they can lead to extreme short term PM$_{2.5}$ concentrations and these are expected to increase in the future (Preisler et al., 2015). In addition to the large PM$_{2.5}$ impacts, wildfire plumes can enhance ozone levels although the emissions remain very uncertain (Cai et al., 2016). A yearlong study in 2015 estimated that forest fires contributed to 12% of organic carbon in PM$_{2.5}$ on an annual basis (Bae et al., 2019).

In this study, we analyze the long term trends in hourly mole fractions of nitrogen oxides (NO$_x$) and daily concentrations of speciated PM$_{2.5}$ from 2000 to 2017. Hourly concentrations of PM$_{2.5}$ were also analyzed from 2012 to 2017. Multiple linear regression was used to identify the impact of meteorological factors such as wind speed, wind direction, temperature, humidity and mixing height. Furthermore, the regression method was used to separate the impacts of variations on the time scale of years, seasons, day of week and time of day. In particular, diurnal profiles are used to yield insights into pollution sources and atmospheric processing. NO$_x$ profiles show the changes in emissions due to day-of-week patterns in vehicle miles traveled, and PM$_{2.5}$ profiles show the impact of weekend and holiday effects.

Methods
Measurements
Measurements were made at the Fresno-Garland super-site in California, which has been at location (36.7853°N, 119.7742°W) since 2012 and was at (36.7813°N, 119.7732°W) before that time. The site is in a residential neighborhood approximately 8 km north of downtown Fresno. It is 1.5 km east of a commercial district located along Highway 41. Skiles et al. (2018) further describe the site location.

Hourly measurements of nitrogen oxides (NO$_x$) and ozone (O$_3$) were obtained from the US EPA Air Quality Datamart (US Environmental Protection Agency, 2018) for 2000 to 2017. Hourly PM$_{2.5}$ concentrations were available for 2012 to 2017. Measurements were linearly interpolated during gaps of 4 hours or less. Of the 18 years (157,800 hours) time series, NO$_x$ was available 97.1% of the time and ozone 98.9% of the time. For PM$_{2.5}$ data were available 96.7% of the time during the 6 years (52,608 hours). Daily averages were obtained from midnight to midnight in Pacific Standard Time (PST, 8 hours behind UTC). Data availability and average concentrations are shown in Table 1.

Uncertainties in the NO$_x$ measurements were discussed in de Foy (2018); Lamsal et al. (2015); Dunlea et al. (2007),
Table 1: Statistical metrics for the hourly multiple linear regression (MLR) models for NO\textsubscript{x} and PM\textsubscript{2.5}. Correlation coefficient squared ($r^2$) shown using transformed variables and excluding outliers ($r^2$ IRLS) and with all untransformed data ($r^2$ All). Measurement averages shown for NO\textsubscript{x} in ppb and PM\textsubscript{2.5} in $\mu$g/m$^3$. DOI: https://doi.org/10.1525/elementa.384.t1

| Species | Date Range          | No. Data | No. Outliers | $r^2$ IRLS | $r^2$ All | Data Avail. (%) | Average |
|---------|---------------------|----------|--------------|------------|-----------|----------------|---------|
| Hourly, Winter |                       |          |              |            |           |                |         |
| NO\textsubscript{x} | Jan 2000–Dec 2017   | 49,986   | 2,417        | 0.82       | 0.58      | 96.2           | 47.4    |
| PM\textsubscript{2.5} | Nov 2011–Feb 2014   | 8,533    | 397          | 0.68       | 0.55      | 98.6           | 31.0    |
| PM\textsubscript{2.5} | Nov 2014–Feb 2017   | 8,509    | 407          | 0.68       | 0.47      | 98.3           | 30.1    |
| Hourly, Summer |                      |          |              |            |           |                |         |
| NO\textsubscript{x} | May 2000–Sep 2017   | 64,525   | 3,427        | 0.86       | 0.76      | 97.6           | 13.2    |
| PM\textsubscript{2.5} | May 2012–Sep 2014   | 10,061   | 397          | 0.42       | 0.29      | 91.3           | 10.4    |
| PM\textsubscript{2.5} | May 2015–Sep 2017   | 10,958   | 517          | 0.48       | 0.33      | 99.5           | 9.7     |

and include interference from other non-NO\textsubscript{x} reactive nitrogen compounds, especially in the afternoons. These should be kept in mind when evaluating the results for NO\textsubscript{x}, but their impacts are mitigated because the Fresno site is an urban site that is strongly impacted by local emissions and can hence be expected to have low fractions of NO\textsubscript{x} species.

Daily measurements of PM\textsubscript{2.5} mass concentration and speciation were obtained from the Chemical Speciation Network (Solomon et al., 2014). The PM\textsubscript{2.5} concentrations were available for 94% of the days. Speciation measurements were carried out every third day. Malm et al. (2011) describe the scaling factors used to convert concentrations of speciated fractions to obtain components of a reconstructed PM\textsubscript{2.5} time series that would match the PM\textsubscript{2.5} measurements. Elemental and organic carbon were available using the IMPROVE measurement method starting in March 2004 and using the CSN method only prior to that. A continuous time series was constructed for both elemental carbon (EC) and organic carbon using the corrections described in Malm et al. (2011). Particulate organic matter (POM) was obtained by scaling organic carbon by 1.8. Particulate ammonium nitrate (NH\textsubscript{4}NO\textsubscript{3}) and ammonium sulfate ((NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4}) concentrations were obtained by assuming that they are fully neutralized and hence scaling the nitrate fraction by 1.29 and the sulfate fraction by 1.375. The chloride fraction was scaled by 1.8 to get an approximation of sea salt. Soil was obtained by summing aluminum, silicon, calcium, iron and titanium, scaled by 2.2, 2.49, 1.63, 2.42 and 1.94 respectively. The data were available for 31 to 33% of the days, as shown in Table 2. The reconstructed PM\textsubscript{2.5} matched the measurements with an $r^2$ of 0.95, as shown in Figs. S1 and S2.

The most complete set of meteorological observations including wind speed and direction, temperature, humidity and pressure were made at the Fresno Yosemite International Airport (KFAT) and were obtained from the Integrated Surface Hourly Database (ISHD) from the National Climatic Data Center (NCDC). The airport is located 5 km east of the measurement site. As with the AQS data, linear interpolation was used to fill gaps of 4 or fewer hours.

**Boundary layer heights**

The mixing height is a key input to the multiple linear regression model. We used boundary layer heights from the fifth generation atmospheric reanalysis of the European Centre for Medium-Range Weather Forecasts, ERA5 (Copernicus Climate Change Service (C3S), 2017). The model has a horizontal resolution of 31 km. Hourly data were obtained for the grid point closest to the measurement site.

Preliminary tests were also performed using the ERA-Interim dataset (Dee et al., 2011) and simulations using the Weather Research & Forecast model (Skamarock et al., 2005) described in Skiles et al. (2018) and Bae et al. (2019). The ERA-Interim heights were available every 3 hours and were interpolated to hourly resolution. This led to poorer model fits near sunrise and sunset. The WRF simulations were at hourly resolution and 9 km horizontal grid resolution, but did not yield regression models with improved correlation coefficients. Overall, it was found that the ERA5 estimates yielded the multiple linear regression model with the best fit with measurements.

**Multiple linear regression model**

Multiple Linear Regression (MLR) models were developed for hourly NO\textsubscript{x} mole fractions and PM\textsubscript{2.5} concentrations; and for daily speciated PM\textsubscript{2.5} components. An additional model was developed for O\textsubscript{3}, (O\textsubscript{3} + NO\textsubscript{x}) to compare with the NH\textsubscript{4}NO\textsubscript{3} model. The MLR model was described in detail in de Foy (2018) where it was used to evaluate the trends and diurnal patterns of NO\textsubscript{x} emissions in Chicago. The Chicago study built upon prior use of the model to identify the impact of stratospheric transport on surface ozone concentrations in the Tibetan plateau (Yin et al., 2017), and to analyze trends and patterns in NO\textsubscript{x} columns from OMI satellite retrievals over China (de Foy et al., 2016b) and the USA (de Foy et al., 2016a).
Table 2: Statistical metrics for the 24-hour multiple linear regression (MLR) models for NO\textsubscript{x}, PM\textsubscript{2.5}, and speciated PM\textsubscript{2.5} components from 2000 to 2017 inclusive. Correlation coefficient squared ($r^2$) shown using transformed variables and excluding outliers ($r^2$ IRLS) and with all untransformed data ($r^2$ All). Measurement averages shown for 2000–2017, 2000–2002 and 2015–2017 along with the annual trend from the measurements and the MLR model. Concentrations of aerosols are in $\mu g/m^3$; mole fractions of NO\textsubscript{x} are in ppb. DOI: https://doi.org/10.1525/elementa.384.t2

| Species          | No. Data | No. Outliers | $r^2$ IRLS | $r^2$ All | Data Avail. (%) | 2000–2017 Average | 2000–2002 Average | 2015–2017 Average | 24-hr Av Trend (%/yr) | MLR Trend (%/yr) |
|------------------|----------|--------------|------------|-----------|----------------|-------------------|-------------------|-------------------|---------------------|------------------|
| Daily, Winter    |          |              |            |           |                |                   |                   |                   |                     |                  |
| NO\textsubscript{x} | 2,106    | 114          | 0.88       | 0.81      | 97.3           | 47.69             | 66.78             | 31.11             | -4.25               | -5.32            |
| PM\textsubscript{2.5} | 2,042    | 113          | 0.80       | 0.61      | 94.3           | 29.56             | 40.72             | 22.07             | -3.20               | -2.90            |
| EC               | 690      | 31           | 0.85       | 0.75      | 31.9           | 1.64              | 2.27              | 1.03              | -4.87               | -5.08            |
| POM              | 690      | 36           | 0.85       | 0.79      | 31.9           | 12.80             | 24.41             | 8.68              | -6.55               | -6.08            |
| NH\textsubscript{4}NO\textsubscript{3} | 678      | 39           | 0.75       | 0.42      | 31.3           | 12.70             | 16.24             | 8.93              | -2.81               | -4.34            |
| (NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4} | 678      | 37           | 0.71       | 0.54      | 31.3           | 1.64              | 2.05              | 1.27              | -3.32               | -3.19            |
| Soil             | 687      | 30           | 0.83       | 0.66      | 31.7           | 0.65              | 0.56              | 0.58              | 1.84                | -0.29            |
| Sea Salt         | 690      | 44           | 0.65       | 0.40      | 31.9           | 0.43              | 0.31              | 0.39              | 0.89                | 2.74             |
| Daily, Summer    |          |              |            |           |                |                   |                   |                   |                     |                  |
| NO\textsubscript{x} | 2,726    | 148          | 0.92       | 0.84      | 99.0           | 13.16             | 19.61             | 7.35              | -6.38               | -6.49            |
| PM\textsubscript{2.5} | 2,574    | 98           | 0.67       | 0.34      | 93.5           | 10.19             | 11.24             | 9.66              | -0.92               | 0.01             |
| EC               | 903      | 42           | 0.88       | 0.75      | 32.8           | 0.52              | 0.66              | 0.31              | -5.59               | -5.64            |
| POM              | 903      | 45           | 0.75       | 0.37      | 32.8           | 4.71              | 5.15              | 4.69              | -1.62               | -0.73            |
| NH\textsubscript{4}NO\textsubscript{3} | 853      | 45           | 0.63       | 0.54      | 31.0           | 1.65              | 2.31              | 1.03              | -5.53               | -4.36            |
| (NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4} | 853      | 32           | 0.63       | 0.29      | 31.0           | 1.97              | 2.39              | 1.42              | -3.79               | -3.76            |
| Soil             | 910      | 54           | 0.81       | 0.64      | 33.0           | 1.41              | 1.17              | 1.58              | 2.49                | 1.70             |
| Sea Salt         | 910      | 54           | 0.34       | 0.04      | 33.0           | 0.13              | 0.03              | 0.18              | 8.14                | 1.02             |

The MLR models are described by Equation 1:

$$
\text{log(CONC + Offset)} = \sum_{y=2000}^{2017} \alpha_y t_y + \sum_{i=1}^{24} \alpha_{i, \text{mon}} t_{\text{mon}, i} + \sum_{wkt=1}^{11} \alpha_{wkt, \text{mon}} t_{\text{mon}, wkt} + \sum_{i=1}^{24} \sum_{wkt=1}^{11} \alpha_{wkt, i, \text{mon}} t_{\text{mon}, wkt, i} + \sum_{j=1}^{5} \alpha_{PBLH, j} t_{PBLH, j} + \sum_{j=1}^{5} \sum_{i=1}^{24} \alpha_{WS, i, j} t_{WS, i, j} + \text{Optimized Inputs} + \alpha_{\text{pdeps}} + \epsilon \tag{1}
$$

Where CONC is the concentration of the species which is being modeled and the offset is used to improve the log-normal fit of the transformed variable: for NO\textsubscript{x}, O\textsubscript{3} and speciated PM\textsubscript{2.5} components it is 0, and for PM\textsubscript{2.5} it is 5 $\mu g/m^3$.

Time vectors $t$ vary between 0 and 1 and are used to represent different temporal terms. For example, $t_y$ represents the variation from year to year. This means that $t_{2011}$ is 1 during 2011 and 0 otherwise. Seasonal variations were represented using a factor $t_{\text{mon}, i}$ for each month of the year. Weekday variations are represented by $t_{\text{mon}, wkt}$, with one vector for each day of the week and one or two extra vectors for the holidays.

MLR models for the hourly measurements used $t_{\text{mon}, wkt, i}$ for each hour of the day for different days of the week to represent the diurnal variations. Separate diurnal factors were used for Mondays, midweek (Tuesdays to Thursdays), Fridays, Saturdays, Sundays and holidays (see the section on the holidays for more details), for a total of 6 sets of 24 factors. Daily models did not include the hourly time factors.

The impact of winds and mixing heights on the models is expected to be non-linear, and were therefore included in the model as a generalized additive model with multiple factors to approximate the non-linear relationship. This was implemented by separating the data into quartiles and by including a regression factor for each of the 5 points separating the quartiles (0%, 25%, 50%, 75% and 100%). For the boundary layer height, there are 5 time series represented by $t_{PBLH, j}$. Piecewise linear interpolation was used to give them a value of 1 at the lower edge of the quartile which then decreases to 0 by the next quartile. As an example, $t_{PBLH, 1}$ is 1 for the times when the mixing height is at the data minimum and it is 0 when the mixing height is above the first quartile. At times when the mixing height is between these two levels, $t_{PBLH, j}$ varies linearly. For the winds we use a two-dimensional array of factors with 5 factors for windspeed and 8 factors for hourly wind direction. As an example, $t_{WS, 1, i}$ represents times when the winds speeds are in the bottom quartile ($i = 1$) and when
the wind direction is from the north \((j = 1)\). This yields 40 factors representing wind speed and direction for the hourly model. The number of factors for wind direction was reduced to 4 for the daily averaged winds, to make sure that each factor had a sufficient number of non-negative values to avoid overfitting in the regression model.

Because there is a large difference in the air quality issues during the winter and the summer months as noted in the introduction, the MLR models were carried out separately according to the seasons. By considering the variations in concentrations of the different pollutants, it was decided to classify November, December, January and February as winter months and May through September as summer months. March, April and October were deemed to be transition months and were not included in the analysis.

The “Optimized Inputs” in Equation 1 represent the meteorological variables selected as inputs to the MLR model for each species and for each season. An optimization algorithm was used to test all candidate variables as an input to the MLR model one at a time, and included the one contributing to the greatest increase in the correlation coefficient. This was carried out iteratively as long as the new input variable contributed to an increase in \(R^2\) of at least 0.005.

Meteorological parameters were used as candidate inputs for all the models. The list of candidate input variables included temperature \((T)\), specific humidity \((q)\), relative humidity \((RH)\), 24-hour precipitation and surface pressure. Fog was included in the model as a time series of the fraction of hours in the preceding 24 hours that were foggy. For rain, a similar time series as fog with the fraction of hours in the preceding 24 hours that were foggy. For rain, a similar time series as fog with the fraction of hours experiencing precipitation was used.

For the hourly model, 24-hour running averages were used of all the meteorological parameters except for winds, boundary layer heights and ozone mole fractions. This was done so that the diurnal profile of the MLR model would be closer to the diurnal profile of emissions and production, as discussed in de Foy (2018). For the daily model, daily averages, minima and maxima were used as candidate inputs to the model. All variables included in the regression model were first normalized.

Finally, hourly ozone was included as an input for the \(NO_2\) model using log transformation \((\log(O_3 + 10))\). As discussed in de Foy (2018), this serves as an approximate proxy for the lifetime of \(NO_3\); high ozone mole fractions are associated with a more reactive atmosphere and hence shorter \(NO_3\) lifetimes.

The baseline term is included as \(\alpha_{\text{baseline}}\) and represents a model-derived average concentration excluding the impact of the factors in the model. Meteorological factors such as temperature and humidity are included as normalized factors with zero mean and hence their impact on the baseline relative to the average concentration is close to 0. The other factors in the model are based on time series of 0's and 1's, and hence can have a net contribution to the overall MLR time series, depending on the value of the MLR coefficient. In practice, the temporal factors within each group sum to 0 because of the weighting on the MLR coefficients described below. Therefore the annual factors, the weekday factors and the hourly factors each have zero net contribution to the average concentration. The MLR baselines were found to be lower than the measured average concentrations mostly because of the impact of the wind and mixing height factors.

The residual between the regression model and the measurement time series is \(\epsilon\).

Table 3 shows the optimized set of variables selected for each model. As an example, Equation 2 shows the optimized inputs for the \(NO_2\) hourly model during the winter months which uses ozone and specific humidity \((q')\) is linearly transformed specific humidity).

\[
\text{Optimized Inputs (}NO_2, \text{Winter, hourly}) = \alpha_{\text{baseline}} \log(O_3 + 10) + \alpha_q q'
\]

There are regression terms for each year, each month, each day of the week and each hour of the day in the time series. This means that the matrix containing all the terms is singular: it does not have a unique solution. This problem is solved by including the magnitude of the regression coefficients as a cost term in the least square analysis as is done routinely in inverse simulations (de Foy et al., 2015). A weighting factor of 1 was used on the penalty term for the regression coefficients except for \(\alpha_{\text{baseline}}\). Using weighting factors has a negligible impact on the results but forces the regression coefficients \(\alpha\) to have the smallest possible values.

The model is solved by constructing a matrix with a column for each time series used as an input and a vector of regression coefficients \(\alpha\). The weighting terms on the coefficients are included as extra rows in the matrix. The coefficients are obtained with a least square inversion of the matrix. Overall, the daily models have between 62 and 76 free parameters to be optimized using between 637 and 2,749 data points. The hourly models for \(NO_2\) have between 223 free parameters to be optimized using between 49,986 and 64,525 data points. The hourly models for \(PM_{2.5}\) have between 209 and 212 free parameters to be optimized with around 10,000 data points for the summer models and around 8,500 for the winter models for the simulations before and after the introduction of the Check Before You Burn program. In all cases the number of data points greatly exceeds the number of free parameters in the model.

There are three ways of interpreting the regression coefficients will be used. The first is as a relative difference from the baseline concentration, as shown in Equation 3 (see de Foy et al., 2016a). The exponential term accounts for the log transformation of the data. For normalized variables, the coefficient \(p\) corresponds to the percentage increase due to a one standard deviation increase in the variable. The temporal time series \((\text{eg. } t_{\text{wkd}}, t_{\text{hr}}, t_{\text{yr}}, t_{\text{m}}, t_{\text{d}}, t_{\text{u}})\) have a value of 1 during the time interval and 0 otherwise. In these cases, the coefficient \(p\) corresponds to the percentage increase in the concentration during those time intervals. The second way to interpret the coefficients is to convert them to a difference in concentration relative to the baseline level using Equation 4. Finally, meteorologically and temporally-adjusted concentrations can be obtained using Equation 5. These are useful for obtaining a trend of concentrations, for example by time of day or by year,
after accounting for the variability introduced by all the other variables in the regression model.

\[ p = (e^a - 1) \times 100\% \]  
\[ \Delta C = (e^a - 1)e^{0.0015} \]  
\[ C_{\text{adjusted}} = e^a e^{0.0015} - \text{Offset} \]

Since the results of least squares methods are sensitive to outliers, an Iteratively Reweighted Least Squares (IRLS) procedure was used to screen them out. Measurement times when the model residual was greater than two standard deviations of all the residuals were excluded from the analysis. This was repeated iteratively until the method converged on a stable set of outliers.

**Holidays**

The standard set of holidays used by the SMOKE model of the US Environmental Protection Agency is New Year's day, Good Friday, Memorial Day, July 4th, Labor Day, Thanksgiving, Black Friday, Christmas Eve and Christmas Day. In addition, July 3rd or 5th are added in the years when they are observed as a holiday. A separate coefficient was used in the MLR models to estimate the change in concentrations on all holidays compared with the days of the week. The residual term (\( e \)) was then analyzed for the individual days to estimate the variability between them. This showed that Good Friday and Christmas Eve resembled regular week days more than the holidays. Furthermore, Black Friday and July 4th were significantly different from the other holidays: Black Friday because of the changed traffic pattern and July 4th because of evening emissions. New Year’s eve was tested as a possible extra holiday but was not found to behave like a holiday.

Based on the data analysis results, it was decided to remove Good Friday and Christmas Eve from the list of holidays and to treat them like regular weekdays. For PM\(_{2.5}\), all other holidays listed above were included in a single category. For hourly NO\(_x\) there were enough data points to include separate factors for Black Friday and July 4th.

## Results and discussion

**Time series and regression metrics**

Figure 1 shows the 18 year time series of daily average mole fractions for NO\(_x\) and PM\(_{2.5}\) concentration. Both the measurement time series and the multiple linear regression model fit are shown. From this, it is clear that both NO\(_x\) and PM\(_{2.5}\) concentrations have decreased over the years. Boxplots of the time series are shown by year, by day of the week, and by time of day in the supplemental material for reference.
**Figure 2** shows the daily average concentrations of selected speciated components of PM$_{2.5}$: elemental carbon, POM, ammonium nitrate and ammonium sulfate. Elemental carbon has followed a similar trend to NO$_x$ with much reduced concentrations, which are particularly noticeable during the winter. POM concentrations are much higher than elemental carbon, but these too have been decreasing. As expected, ammonium nitrate occurs

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**Figure 1:** Time series of daily averaged NO$_x$ and PM$_{2.5}$ at the Fresno Supersite along with the optimal multiple linear regression (MLR) models and model outliers. Crosses represent times with concentrations above the axis, circles are points rejected as outliers by the Iteratively Reweighted Least Squares (IRLS) algorithm. DOI: https://doi.org/10.1525/elementa.384.f1

**Figure 2:** Time series of Chemical Speciation Network PM$_{2.5}$ concentrations for elemental carbon, particulate organic matter, ammonium nitrate and ammonium sulfate of PM$_{2.5}$ along with the optimal multiple linear regression model and model outliers. Crosses represent times with concentrations above the axis, circles are points rejected as outliers by the Iteratively Reweighted Least Squares (IRLS) algorithm. DOI: https://doi.org/10.1525/elementa.384.f2
nearly exclusively in the winter and has been going down considerably. Ammonium sulfate levels are very low with much less seasonal variation but a significant decrease in concentrations over the years.

Table 2 shows the average concentrations over the entire time series as well as for the first three years and the last three years of data. The average hourly mole fraction of NOx is 47.7 ppb in the winter and 13.2 ppb in the summer, and for PM<sub>2.5</sub> the average concentration is 29.6 μg/m<sup>3</sup> in the winter and 10.2 μg/m<sup>3</sup> in the summer. Fresno County is in non-attainment of the National Ambient Air Quality Standards for PM<sub>2.5</sub>.

The statistical metrics of the multiple linear regression models are shown for the winter and summer simulations in Table 1 for the hourly models and Table 2 for the daily models. The multiple linear regression model performed best in the summer for NOx with R<sup>2</sup> coefficients of 0.86 for the transformed variables excluding outliers. The model assumes that the temporal effects do not vary significantly throughout the simulation time period. This was found to be a valid approximation for most of the models, as shown in Figs. S3 and S4 for NOx. Hourly PM<sub>2.5</sub> models however were found to differ before and after the introduction of the Check Before You Burn program and hence have separate models, as shown in Table 1.

The daily PM<sub>2.5</sub> concentrations are from the CSN network and are available from 2000 to 2017. The R<sup>2</sup> are somewhat higher for the daily models than for the hourly models, ranging from 0.63 to 0.92 for the transformed data without outliers. This is mostly because of the much larger number of data points in the hourly model than in the daily model. It also suggests that it is more difficult to simulate the diurnal profiles of aerosols than either the NOx diurnal profiles or the other temporal scales of aerosol concentrations. The exception to the high R<sup>2</sup> is the sea salt model that performed poorly in the summer. Both soil and sea salt have much lower concentrations than the other PM<sub>2.5</sub> components and will not be discussed further in the present work.

Uncertainty in the MLR coefficients was analyzed using block-bootstrapping (de Foy, 2018). Figs. S5–S8 show the probability density functions of NOx and PM<sub>2.5</sub> measurements by weekday, along with the uncertainty in the bootstrapped mean and the uncertainty in the MLR coefficients. Overall, the figures show that the MLR method reduces the uncertainty in the estimates of the weekday temporal profile compared with analysis of data means.

Meteorological factors

Table 3 shows the variables included in each model. All hourly models include the hourly ERA5 boundary layer heights and the wind speed and direction observed at the airport. After winds and boundary layer height, humidity was the most frequent predictor either as relative humidity or specific humidity mostly for the winter months. Specific humidity higher than 1 standard deviation is associated with a 17% drop in NOx during the winter, but has no effect on NOx in the Summer. For PM<sub>2.5</sub>, relative humidity is associated with 17% drop in concentrations in the winter and 6% increase in the summer. Temperature is associated with 18% higher PM<sub>2.5</sub> in the summer. Rain is associated with 68% lower PM<sub>2.5</sub> in the winter. Although fog only contributed to the PM<sub>2.5</sub> model in winter, it was associated with an increase in concentration of 125%.

For the daily models, average daily winds contributed to all the models. The optimum boundary layer heights were usually the daily minimum from ERA5. In the case of NOx, this is most likely because the highest mole fractions occur during the morning rush hour and contribute the most to the daily average. The models for PM<sub>2.5</sub> in the winter and the summer include both daily minimum and daily average heights. This is most likely because PM<sub>2.5</sub> experiences high values as a primary species in the morning and as a secondary species in the afternoon.

Most of the daily models used temperature as an input, often relying on both minimum and maximum daily temperature, and many of the models also used humidity, usually as relative humidity. Average surface pressure was associated with small contributions to summer PM<sub>2.5</sub>. In the winter, fog was associated with increases of 132% for PM<sub>2.5</sub>, 620% for NH₄NO₃ and 400% for (NH₄)₂SO₄. Fog occurs infrequently, and hence individual episodes can have a larger impact on the MLR coefficients than smoothly varying inputs such as temperature and humidity. Nevertheless, the increases are consistent with enhanced particle growth measured on high-fog days (Betha et al., 2018). Reduced fog frequency was found to be correlated with reduced pollution levels since the passage of the Clean Air Act (Gray et al., 2019), which is consistent with the connection in the MLR model between aerosols and fog events. Enhanced wet deposition of nitrates during fog episodes was found to be offset by enhanced production in surface layers with high relative humidity (Lillis et al., 1999). Measurements of fog water in Fresno confirmed the presence of large amounts of nitrates (Herckes et al., 2007). While the present analysis has identified that higher aerosol concentrations are associated with fog, it cannot determine the causal direction: it is possible that the higher concentrations are a function of the particularly stable and calm wind conditions that lead to fog formation in the first place.

Rain was not selected for the optimal set of variables for daily PM<sub>2.5</sub> but was associated with a decrease of 57% in winter (NH₄)₂SO₄.

**Annual and weekday trends of NOx and PM<sub>2.5</sub>**

Figure 3 shows the annual trends and weekday patterns in mole fraction differences of NOx and concentrations of PM<sub>2.5</sub> for the winter and summer months obtained from the daily MLR models. Table 2 shows the average trend from 2000 to 2017 using a line of best fit for the seasonal averages as well as using the MLR annual factors.

The biggest decrease has been in NOx mole fractions, with reductions of 22 ppb in the winter and 12.5 ppb in the summer which correspond to reductions of more than 50%. There was a sharper drop after 2007 and an uptick in 2012. Although it is too early to tell, it seems that reductions may have plateaued or are possibly increasing in the last 3 years (Jiang et al., 2018). Overall, NOx has been decreasing by around 5 to 6% per year.
During the winter months, the PM$_{2.5}$ trend has tracked the NO$_x$ trend but with a smaller amplitude. Overall the reduction is around 40% since 2000, corresponding to a decrease of around 3% per year. The decrease is less monotonic than that of NO$_x$, with two spikes of 10% increases: one in 2007 and one from 2013 to 2014. During the summer months, there has been very little change in PM$_{2.5}$ over the years. Individual years vary from the baseline by less than 10%. The largest increase was from 2012 to 2013 with a jump of over 10%, followed by a gradual decrease back to baseline levels by 2017.

The weekday pattern is shown in Figure 3 as well as in Table 4. The day-of-week pattern of NO$_x$ factors exhibits a strong weekend effect year-round: mole fractions are 15 to 18% below the midweek baseline on Saturdays, and 26 to 30% below on Sundays and holidays. On Mondays levels are 10% below during the winter and 5% below during the summer. As there is no indication of lower emissions on Mondays compared with other weekdays, this could be the effect of carryover from one day to the next.

PM$_{2.5}$ has a day-of-week pattern that is different from NO$_x$ and also different in the winter compared with the summer. In winter the mole fractions increase gradually throughout the week reaching a maximum of 6% above the baseline on Saturdays and 7% above on holidays. Sundays are only 3% above the baseline and Mondays are 5% below. This suggests that lower emissions on Sundays are offset by the higher emissions on Saturdays, but that they lead to lower mole fractions on Mondays. During the summer there is very little variation throughout the week. Saturdays are similar to the rest of the week, holidays are

| Air Pollutant | Sat | Sun | Hol | Sat | Sun | Hol |
|---------------|-----|-----|-----|-----|-----|-----|
| November–February | May–September |
| NO$_x$ | -17.6 | -25.9 | -25.9 | -15.4 | -30.2 | -28.3 |
| PM$_{2.5}$ | 6.0 | 3.1 | 6.8 | -1.2 | -4.8 | 3.0 |
| EC | 2.4 | -2.1 | 23.8 | -10.2 | -33.2 | -26.6 |
| POM | 11.9 | 15.8 | 34.1 | -0.1 | -6.1 | -8.3 |
| NH$_4$NO$_3$ | -0.9 | -12.7 | -8.8 | -8.8 | -20.3 | 1.9 |
| (NH$_4$)$_2$SO$_4$ | -5.9 | -5.4 | 15.6 | -0.9 | -5.6 | 10.0 |
| Soil | -14.4 | -26.4 | -35.1 | -20.7 | -37.1 | -12.1 |
| Sea Salt | 6.7 | 12.4 | 55.3 | 2.8 | 4.1 | 28.2 |

**Figure 3:** Annual trends (top) and day of week patterns (bottom) in difference in mole fractions of 24-hour average NO$_x$ and concentrations of PM$_{2.5}$ ($\Delta C$) in Fresno, CA using the multiple linear regression model. Winter months shown on the left, summer months on the right, holidays (“Hol”) are defined in the text. DOI: https://doi.org/10.1525/elementa.384.f3
3% higher and Sundays are 5% below. Mondays are 4% below, again suggesting that there is some effect from reduced carryover.

**Diurnal profiles of NOx**

**Figure 4** shows the diurnal profiles for NO\textsubscript{x} for the winter and summer months as adjusted mole fractions, see Equation 5. Separate profiles were calculated for Mondays, midweek (Tuesdays to Thursdays), Fridays, Saturdays, Sundays and holidays. For NO\textsubscript{x} there were enough data points to be able to treat Black Friday and July 4\textsuperscript{th} separately from the other holidays (“Hol”).

The MLR models take into account hourly boundary layer heights and wind speed and direction which means that the models include the effects of vertical mixing and horizontal ventilation. For NO\textsubscript{x}, the effects of ozone are also included which serves as a rough proxy for the chemical lifetime. The diurnal coefficients should therefore be closer than the diurnal profile of mole fractions to estimates of emissions (de Foy, 2018). For comparison, boxplots of the annual trends, weekday profile and diurnal variations of the mole fractions are shown in the supplemental material.

During the winter months, the diurnal scaling factors for NO\textsubscript{x} show the expected increase in emissions starting at 6am and peaking at 9am. There is then a further increase starting at lunch into the early afternoon. The factors then decrease reaching an evening plateau from 6pm to 10pm. Mondays through Fridays have very similar profiles. Saturdays have a slower and more gradual increase reaching a peak at 2pm and experiencing a secondary peak around 8pm. Sundays and holidays are similar to each other with much reduced factors and a delayed and smoother increase in the morning.

During the summer months, NO\textsubscript{x} increases continuously from 6am to 9am in Pacific Daylight Time, which corresponds to 5am to 8am in Pacific Standard Time. The factors then decrease during the day and experience a secondary peak at around 9pm (PDT). Saturdays, Sundays and holidays have delayed and more gradual morning increases and lower levels throughout the day. July 4\textsuperscript{th} however has a large peak starting at 10pm and dropping after 12pm (PDT).

In prior research for Chicago, the MLR coefficients showed a diurnal profile that was consistent with the emissions expected from traffic counts and vehicle speeds (de Foy, 2018). **Figures 5** and 6 show the diurnal profiles of Vehicle Miles Traveled (VMT) for light duty, medium duty and heavy duty vehicles in Fresno county (Eastern Research Group, Inc., 2017). The NO\textsubscript{x} scaling factors from the MLR analysis for the winter months are also shown. A least-squares fit was used to find the optimal combination of the three vehicle classes that would yield the best match to the NO\textsubscript{x} factors. There are large uncertainties associated with trying to obtain an estimate of the contribution of the vehicle classes from their diurnal profiles. Nevertheless, it does provide an estimate based on the analysis of the mole fractions which can serve to compare results from emission inventories.

On Mondays, the fit had an $r^2$ of 0.68 and consisted of 37% light duty, 14% medium duty and 49% heavy duty vehicle emissions. On Fridays the fit is improved, with an $r^2$ of 0.79 and a vehicle emissions mix that is roughly similar albeit with more medium duty and fewer heavy duty emissions (40% of light duty, 22% medium duty and 38% heavy duty). The fit was much better for Saturdays ($r^2$ of 0.88) and Sundays ($r^2$ of 0.85), with a shift to light duty vehicle emissions which made up 65% of the NO\textsubscript{x} factors on Saturdays and 48% on Sundays. This is consistent with a stronger weekend effect for medium and heavy duty vehicles than for light duty vehicles: Light duty VMT drop by 19% on Saturdays and 27% on Sundays; Medium duty VMT drop by 59% and 76%; and heavy duty VMT drop by 45% and 58% respectively (Eastern Research Group, Inc., 2017; Marr and Harley, 2002).

During the summer months there is much more chemical reactivity and the profiles do not match the VMT profiles as well. In particular, the NO\textsubscript{x} scaling factor profiles

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**Figure 4**: Diurnal pattern of adjusted NO\textsubscript{x} mole fractions from the hourly multiple linear regression model by weekday for the winter months (left) and summer months (right) in Fresno, CA. Holidays are defined in the text. DOI: https://doi.org/10.1525/elementa.384.f4
have a large peak in the early morning as well as a secondary peak in the evening. Both peaks correspond to times of the day when the NO\textsubscript{x} lifetime are longer, which suggests that using ozone as a proxy for chemical reactivity in the regression model was not sufficiently accurate. In addition, the peaks could also be due to uncertainties in the vertical mixing height as well as the impact of horizontal air mass transport since wind speeds are higher in the summer months in Fresno.

Figure 5: Top: Normalized diurnal profiles of Vehicle Miles Traveled (VMT) by vehicle class (light duty (LD), medium duty (MD), heavy duty (HD)) for Mondays and Fridays. Bottom: Diurnal profile of winter NO\textsubscript{x} scale factors from the Multiple Linear Regression (MLR) model; bar chart showing the contribution of each vehicle class to a diurnal profile with the best match to the NO\textsubscript{x} scale factors. The percentage contribution of each vehicle class and the correlation coefficient between the sum of the VMT profiles and the NO\textsubscript{x} scale factors are shown inside the legend. DOI: https://doi.org/10.1525/elementa.384.f5

Figure 6: Top: Normalized diurnal profiles of Vehicle Miles Traveled (VMT) by vehicle class (light duty (LD), medium duty (MD), heavy duty (HD)) for Saturdays and Sundays. Bottom: Diurnal profile of winter NO\textsubscript{x} scale factors from the Multiple Linear Regression (MLR) model; bar chart showing the contribution of each vehicle class to a diurnal profile with the best match to the NO\textsubscript{x} scale factors. The percentage contribution of each vehicle class and the correlation coefficient between the sum of the VMT profiles and the NO\textsubscript{x} scale factors are shown inside the legend. DOI: https://doi.org/10.1525/elementa.384.f6

**Diurnal profiles of PM\textsubscript{2.5}**

Figure 7 shows the diurnal pattern of the adjusted concentrations based on the MLR coefficients for PM\textsubscript{2.5} for the winter and summer months. For the winter, the analysis was carried out separately for the three winters before the introduction of the Check Before You Burn program (2011/2012, 2012/2013 and 2013/2014) and the three winters afterwards (2014/2015, 2015/2016 and 2016/2017). For the summer, the analysis was carried out...
for the three summers from 2012 to 2014, and separately for the three summers from 2015 to 2017. **Table 1** shows the statistical metrics of the separate models.

During the winter months before the introduction of the Check Before You Burn program, PM$_{2.5}$ drops before sunrise to nearly 40% below baseline levels, then rises rapidly to reach a 1 pm peak that is up to 70% above baseline levels on Fridays but only 30% above baseline levels on Sundays. The levels drop after 3 pm to reach a secondary low from 4 to 5 pm and then increase in the evening. Saturdays start off like the weekdays, but then have a higher peak from 9 pm to 12 am. This evening feature is even more striking on the holidays which have higher levels starting at 7 pm that last past midnight. Holidays also have higher levels before sunrise, probably as a consequence of emissions from the preceding evenings. In contrast, Sundays have lower midday peaks than other days and have an evening profile that resembles the other weekdays.

Since the introduction of the Check Before You Burn program in November 2014, the diurnal variation has been reduced: midday levels do not reach above 30%. Whereas the daytime peaks are lower than previous years, the nighttime increase on Saturdays is still as strong as before and is now as strong as the daytime peak. On holidays, the daytime levels are much lower, but the evening peak is higher than before, reaching nearly 60% above the baseline.

During the summer months from 2012 to 2014, PM$_{2.5}$ drops in the early morning followed by a broad daytime increase that peaks around midday. The reduction at sunrise is barely noticeable in the data. Saturdays still have a small evening peak that is distinct from the other days. Levels are lower Sundays, Mondays, and especially holidays with delayed increase in the morning compared with the other weekdays, but less difference in the evening. From 2015 to 2017, the diurnal profiles are more similar between the days leading to a more distinctive pattern overall. There is an almost linear increase between 6 am to 1 pm (PDT) on all days. This increase is followed by a linear decrease into the night. As with the other cases, Saturdays still have higher evening levels. The holidays were the most different, with increases reaching up to 30% above baseline at midnight.

By indentifying differences in concentrations on holiday evenings, the MLR analysis yields clues about the emission sources that could be responsible. The diurnal profiles are indicative of the impact of residential wood burning in the night time boundary layer (Zhang et al., 2016).

**PM$_{2.5}$ speciation trends**

Daily speciated PM$_{2.5}$ measurements from the Chemical Speciation Network provide more detailed insight into the trends and patterns in the hourly AQS data of PM$_{2.5}$. 

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**Figure 7:** Diurnal pattern of adjusted PM$_{2.5}$ concentrations by weekday from the hourly multiple linear regression model for the winter months (left) before (top) and after (bottom) the introduction of Check Before You Burn; and summer months (right) for 2012 to 2014 (top) and 2015 to 2017 (bottom) in Fresno, CA. DOI: https://doi.org/10.1525/elementa.384.f7
Figure 8 shows the annual average concentrations and fraction of PM$_{2.5}$ by speciated component. Table 2 shows the average concentrations as well as the linear trends from the regression analysis, and Table 4 shows the strength of the weekend effect by species.

In the winter, PM$_{2.5}$ concentrations have dropped by around 60% between 2000 and 2016, from concentrations of 41 μg/m$^3$ to 22 μg/m$^3$ in 2017. The main component is particulate organic matter which has had a fluctuating fraction of total PM$_{2.5}$ over the years: it made up around 57% of PM$_{2.5}$ in 2000 and 2001, but only 33% in 2005 and 32% in 2014. In recent years particulate organic matter has increased back to fractions above 40%. The second largest component is ammonium nitrate which made up around 32% of PM$_{2.5}$ in 2000, increasing to a maximum of 54% in 2014 and decreasing in recent years. In terms of concentrations, particulate organic matter has reduced the most starting at around 27 μg/m$^3$ in 2000, and decreasing to around 9 μg/m$^3$ in recent winters. Ammonium nitrate used to vary between 13 and 20 μg/m$^3$ but has recently dropped to a low of 7 μg/m$^3$ for 2016. The remaining components of fine aerosols are ammonium sulfate, soils and sea salt. These make up between 6 to 12% of PM$_{2.5}$ with total concentrations decreasing from a high of 3.5 μg/m$^3$ in winter 2002 to a low of 2.0 μg/m$^3$ in 2016.

In the summer, average PM$_{2.5}$ concentrations have dropped from around 10 to 12 μg/m$^3$ in the earlier years to 9 to 10 μg/m$^3$ recently. There was a peak of 15 μg/m$^3$ in 2008 which, in California, had one of the most devastating wildfire seasons of the time series. POM makes up 45 to 55% of summer PM$_{2.5}$. Ammonium nitrate levels have reduced from around 2 μg/m$^3$ to around 1 μg/m$^3$; they
used to make up 20% of PM$_{2.5}$ but now make up around 10%. Ammonium sulfate levels have been fairly stable between 1.2 and 2.5 μg/m$^3$, with concentrations closer to the upper end of the range at the beginning of the time series and at the lower end of the range by 2017. Soil and sea salt are the only components that have increased over time. Soils used to be around 1 μg/m$^3$ and used to make up 10% of PM$_{2.5}$, they now contribute closer to 2 μg/m$^3$ and 20% of PM$_{2.5}$. Sea salt used to be close to negligible, but now contributes around 0.2 μg/m$^3$ which corresponds to 2 to 3% of PM$_{2.5}$.

The increase in PM$_{2.5}$ in 2013 can be seen in the speciated data for the winter months, but in the summer there is an unexpected discrepancy between the reconstructed PM$_{2.5}$ and the measured PM$_{2.5}$ for 2013 and 2014. Comparison with other sites in the valley would be necessary to identify the cause of this feature.

**Annual and weekday trends of speciated PM$_{2.5}$**

*Figure 9* shows the annual trends and weekday patterns of elemental carbon and POM for winter and summer. The differences were obtained using Equation 3: for the annual plots, the average over all years is 0, whereas for the weekday plots 0 is set as the midweek average (Tuesday to Thursday). From the figure, it can be clearly seen that in both the winter and the summer elemental carbon follows the annual trends of NO$_x$ with the exception of a peak in 2005 and 2006. The weekday profile in the summer shows the same pattern, with elemental carbon reduced by 30 to 35% on weekends. During the winter however elemental carbon stays similar to weekdays on Saturdays and Sundays and increases by nearly 30% on holidays. This suggests that elemental carbon in Fresno is mostly associated with NO$_x$ emissions, and hence mostly mobile sources including diesel engines (McDonald et al., 2015).

The strong reductions are consistent with the long term reductions found using haze measurements (Kirchstetter et al., 2017), and the temporal profiles suggest that diesel engines are still the dominant source in the summer. During the winter however, nighttime emissions can contribute very significant levels as well. The temporal profile suggests that these are mostly due to residential wood burning, in agreement with measurements made using aerosol mass spectrometry (Betha et al., 2018).

In the winter, POM follows the same pattern as elemental carbon, with a strong downward annual trend that even exceeds the reductions in NO$_x$. In the summer there is much more variability from year to year than in the winter. A general decrease in levels to 2014 has been offset by a rise for 2015–2017. The day of week profile follows that of PM$_{2.5}$.
with little variation throughout the week, although POM is lower on Mondays by 10% and on holidays by around 15%. This suggests that there is interannual variability most likely from wildfires but also a significant contribution from primary sources in the winter and secondary sources in the summer. The temporal profiles suggest that the winter primary source is residential wood burning, in agreement with measurements (Betha et al., 2018), and that the summer is strongly influenced by secondary organic aerosols, in agreement with modeling studies (Zhu et al., 2019).

Figure 10 shows the annual trends and weekday patterns of ammonium nitrate and ammonium sulfate. In the winter, both ammonium nitrate and ammonium sulfate trends are similar to the overall PM$_{2.5}$ trends with reductions of 4.3% per year for ammonium nitrate and 3.2% per year for ammonium sulfate. The weekday profile of ammonium nitrate is very distinctive with a sharp peak on Thursdays that is 15% above baseline levels. The weekend effect is much smaller than for NO$_x$: levels are down by 1% on Saturdays, 13% on Sundays and 9% on holidays.

In the summer, the concentrations of ammonium nitrate and ammonium sulfate are similar, with 3 year averages of 2.3 and 2.4 μg/m$^3$ at the beginning and 1.0 and 1.4 μg/m$^3$ at the end of the time series. Consequently, the annual trends of ammonium nitrate and ammonium sulfate are similar to each other and to the winter trends: Ammonium nitrate decreased by 4.4% per year and ammonium sulfate by 3.8% per year. These trends show that controls on NO$_x$ have been effective at reducing PM$_{2.5}$ concentrations as discussed by Chen et al. (2014) and Pusede et al. (2016). Note however that in recent years the reductions have been slowing down. This is consistent with a recent slowing down in NO$_x$ reductions (Jiang et al., 2018) and could also indicate a greater importance of agricultural soil NO$_x$ emissions (Almaraz et al., 2018).

Ammonium sulfate does not vary much during the week (not shown): it follows the general profile of PM$_{2.5}$ except for holidays which are 10% higher than the middle of the week (compared with 3% higher for PM$_{2.5}$). This is consistent with ammonium sulfate being part of the regional PM$_{2.5}$ plume, with added concentrations possibly due to holiday activity.

Figure 10 shows the weekday pattern of NH$_4$NO$_3$ in units of μg/m$^3$ versus the pattern of O$_x$ in units of ppb for the winter and summer months. There is a clear correspondence in the patterns for the weekdays. A line of best fit between NH$_4$NO$_3$ and O$_x$ shows that an extra 1 ppb of O$_x$ corresponds to an extra 0.75 μg/m$^3$ of NH$_4$NO$_3$ in the

![Figure 10](https://doi.org/10.1525/elementa.384.f10)
winter months, and to an extra 0.16 μg/m³ in the summer months. This is to be expected as NH₄NO₃ partitions to the particle phase at low temperatures, but remains in the gas phase at higher temperatures.

On winter weekends, NH₄NO₃ decreases by only 0.18 μg/m³ on Sundays compared with a decrease in O₃ of 0.9 ppb. Winds are very weak in the winters and stagnation more prevalent such that the higher NH₄NO₃ concentrations than expected could be due to the presence of a regional plume and the result of day-to-day carryover. In contrast, summer weekends follow the weekday trends, with reduced NH₄NO₃ concentrations in line with the O₃ reductions. Winds are stronger leading to more ventilation of the valley such that NH₄NO₃ levels may be more closely coupled to O₃ concentrations. The exception to the summer weekday patterns are the holidays. Inspection of the residuals in the MLR model suggests that this is mostly due to higher concentrations on July 4th which are probably related to fireworks.

**Summary**

Analysis of 18 years of air pollution measurements in Fresno, California, has shown that there have been large improvements in air quality in the city. NOₓ, elemental carbon and ammonium nitrate concentrations have been reduced by around a factor of two since 2000. NOₓ mole fractions are due to primary emissions and have experienced similar reductions in the winter and summer months, as well as reductions of 15 to 30% on weekends. The changes in weekend emissions was shown to match the reduced vehicle miles traveled as well as the reduced diesel fuel fraction during the weekend.

PM₂.₅ concentrations have not reduced as much as NOₓ over the years and do not exhibit a strong reduction during the weekends. The analysis of the speciated components of PM₂.₅ from the Chemical Speciation Network show that the behavior of the reductions varies by component and by time scale. Ammonium nitrate and ammonium sulfate have decreased by a factor of two over the 18 years, mostly in line with NOₓ reductions, despite being the product of emissions from different sources.

The weekday profile reveals the importance of oxidation in the regional polluted air mass and the importance of day-to-day carryover. Although the present study is limited to Fresno, there are other sites with long term time series of speciated PM₂.₅ in both urban and rural sites in the San Joaquin Valley that could be used to further analyze regional effects.

Elemental carbon concentrations have also gone down in line with NOₓ mole fractions over the years, but the day of week and time of day pattern show that residential wood combustion remains an important contributing factor to overall concentrations. Particulate organic matter has gone down during the winter, but not during the summer. Increases in organic carbon during winter weekends corroborate the impact of residential wood combustion seen in the elemental carbon concentrations. Inter-annual variations of organic carbon during the summer months suggest that recent increases in forest fires have offset the reductions that might have been expected from reduced anthropogenic emissions.

**Data Accessibility Statement**

The Air Quality System Data used in this study are available from US Environmental Protection Agency (2018), (http://www.epa.gov/aqs). The meteorological data are available from the US National Climatic Data Center Integrated Surface Database which can be accessed at: http://www.ncdc.noaa.gov/isd. The ERA5 dataset from the European Centre for Medium-Range Weather Forecasts is available through the Climate Data Store of the Copernicus Climate Change Service which is available at https://cds.climate.copernicus.eu. The traffic data are available in the report from Eastern Research Group, Inc. (2017).

**Supplemental file**

The supplemental file for this article can be found as follows:

- Text S1. Supplemental material. DOI: https://doi.org/10.1525/elementa.384.s1

**Acknowledgements**

We thank Allison DenBleyker of Eastern Research Group, Inc., for helpful comments on the manuscript. We are grateful to the anonymous reviewers for their careful reviews which have led to improvements in the manuscript.

**Competing interests**

The authors have no competing interests to declare.

**Author contributions**

- Contributed to conception and design: BdF, JJS
- Contributed to analysis and interpretation of data: BdF, JJS
- Drafted and/or revised the article: BdF, JJS
- Approved the submitted version for publication: BdF, JJS

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