Quantifying and Interpreting Trends in Air Toxics

Are air toxics concentrations changing?

Are the ambient concentration changes in response to changes in emissions?

Trends in Air Toxics

What's Covered in This Section

- This section focuses on trends in ambient air toxics over time; diurnal and seasonal trends are discussed in *Characterizing Air Toxics*, Section 5.
- The following topics are addressed in this section:
  - Quantifying Trends
    - Overview of trends analysis
    - Setting up the data for trend analyses
    - Effect of changes in MDL on trends
    - Summarizing trends
    - Discerning and quantifying trends
      - Quantifying Trends
      - Visualizing Trends
    - Aggregating trends to larger spatial areas
  - Interpreting Trends
    - Evaluating annual trends in the context of control programs
    - *Adjusting trends for meteorology (introductory)*

Trends Overview
Motivation

Assessing trends is useful. Monitoring data are needed to track air toxics concentrations and their changes over time. One of the major programmatic objectives for air toxics measurements is providing data to track progress toward emission and risk-reduction goals. The ability to detect trends in ambient concentrations that are associated with planned air quality control efforts is needed to assess the effectiveness of emission control programs. For example, if specific control strategies have been implemented in an area to reduce emissions of tetrachloroethylene from dry cleaners, do the ambient data indicate that concentrations have decreased since the implementation of the control?

Visual inspection of trends is important. Air quality data typically do not fit a normal distribution. The data tend to be skewed and exhibit a few high concentration events. Thus, trends in extreme values in a data set may differ significantly from trends observed in a statistic that describes the bulk of the data. Different statistical metrics can be examined to look for trends. For example, the annual maximum pollutant concentrations can be plotted to assess how annual peak days are changing over time, or the median concentrations can be plotted to assess how the 50th percentile of the days are changing. In addition, to assess a trend in air quality, representative data are required to estimate a trend that is meaningful.

Understanding the data uncertainties is necessary. Uncertainties impact our ability to clearly discern air quality trends and distinguish between “real” changes and artifacts. For example, measurement accuracy, interferences, and the amount of data above method detection limits, need to be understood to properly interpret the data.

Obtaining consensus (or weight of evidence) among results from different approaches increases our certainty in the observed trends. Quantifying and interpreting trends can be complicated (e.g., there are many different methods). The analyst needs to understand methods for quantifying trends and determining their statistical significance. When several different approaches or “looks” at the data point to the same conclusion, confidence in the conclusion is increased. The analyst also needs to be able to communicate the results in a meaningful and understandable way. Interpretation of trends from site level to larger scales, such as city-wide or regional scale, needs to be done with care. Some site and pollutant combinations may be dominated by local sources or comparisons between some sites may not be reasonable because of large differences between sampling methods.

Trends Overview

Analysis Questions

- Are concentration levels changing at a monitoring site?
- Are changes consistent across sites, areas, or regions?
- Are changes consistent across pollutants or pollutant groups?
- Are changes consistent across time periods?
- Are changes consistent with expectations (e.g., emissions controls, changes in population)?

Setting Up Data for Trend Analysis
Overview

Steps to prepare data for trend analysis:

- Acquire and validate data (covered in *Preparing Data for Analysis,* Section 4)
- Identify and treat data below detection in preparation for annual averages (covered in this section)
- Create valid annual averages or other metrics for trends (*subannual data averaging is covered in Preparing Data for Analysis,* Section 4)
- Create valid site-level trends (covered in this section)

Setting Up Data for Trend Analysis

Identifying Censored Data

- Data are typically reported as a concentration value with an accompanying method detection limit (MDL). In AQS, the MDL is either a default value associated with the analytical method (MDL) or a value assigned by the reporting entity for that specific record (alternate MDL).
- NATTS program guidance suggests that laboratories report all values, regardless of the MDL. However, many air toxics data are reported as censored values; i.e., they have been replaced with zero, MDL/2, or MDL (or some other value).
- Identifying censored values is a helpful first step in treating data below detection. Reporting of censored data will most likely differ among sites and may even be different by method, parameter or time period for a given site. For this reason it is recommended that censored data analyses be carried out for each site, parameter, and method, and temporal variability should be considered.
- Data may be identified and separated at or below the detection limit along with the associated MDL and date/time; if alternate MDLs are available, it is recommended they be used rather than the default MDLs.
- Data may be examined for obvious substitution. Count the number of times each value at or below detection is reported at a given site, parameter, and method. Are the majority of data reported as the same value (e.g., zero or MDL/2)?
  - If data are largely reported as two or more values, investigate the temporal variation of the data. Are there large step changes where reporting methods or MDLs have changed?
  - Do the duplicate values indicate a typical censoring method (e.g., MDL/2, MDL/10)?
  - Alternate MDLs may be different for each sample run causing a distribution of values if MDL/x substitutions were used. Just because values below MDL are not all the same does not mean they are not censored!
- Check for MDL/X substitution.
  - Make a scatter plot of the value vs. MDL to see if the data fall on a straight line.
  - If the data do form a straight line, the slope of the regression line will indicate the value by which the MDL has been divided.
    - Is the value a reasonable number that would be used for MDL substitution (e.g., 1,2,5 or 10)?
      - If the data have been formatted, processed or converted, ratios may not be exactly the same due to rounding differences; the distribution should be close to a straight line and centered around a single integer if MDL/x substitutions have been made.
      - If a bifurcated pattern is observed, the substitution method may have changed over time. Plot a time series of the ratios and look for step changes.
    - The distribution of the ratios should be highly variable if the data are not censored.

Setting Up Data for Trend Analysis
Treating Data Below Detection *(1 of 2)*

- Following are suggested steps to create averages:
  - If uncensored values (i.e., NOT zero, MDL/2, or MDL) are reported below MDL, use the data "as is" with no substitution.
  - If uncensored values are not available, substitute MDL/2 for data below MDL or use more sophisticated methods as described in Section 4.
  - If there is a mix of censored and uncensored data,
    - In data sets with a mixture of censored and uncensored data, two substitution methods can be compared: (1) MDL/2 substitution for censored values and leave uncensored values "as is" and (2) MDL/2 substitution for all data below detection
    - If results are in the same direction using both substitution methods, confidence in the results is increased and substitution method 1 should be retained. If the results do not agree, a more sophisticated method for estimating the data below MDL should be employed.
  - For all data sets, identify the percentage of data below MDL for each year in the trend period. It is important to keep track of how much data are below detection to better understand possible biases in the average. Even if censored values are not used, keep a record of this information to provide one measure of the uncertainty in the results.

Setting Up Data for Trend Analysis

Treating Data Below Detection *(2 of 2)*

- Each annual average should have an associated calculation of the percent below detection. These data provide information about the biases of the annual average when data are below detection.
- When assessing trends over time for a pollutant,
  - Assess trends at all sites regardless of the percent of data below MDL. Note, however, that data are below detection for many site/pollutant combinations. To avoid over-interpretation of observed trends, it is recommended the trend values and their associated percent below detection be visually inspected. Consider trends at sites where at least half of the years for a given trend period have at least 15% of their measurements above MDL for that year.
- For the national level analyses, a 15% “cut-off” was selected based on review of a small data set with most data above detection. Bias in the annual average was investigated for this data set across a range of percent of data below detection. At 15% below detection, the bias in the annual average was 10-40%. A more stringent cut-off may be required if less bias is desirable.
  - For example, if a 5% concentration change was observed but all years have greater than 85% data below detection, the analyst cannot be sure whether this change is real or an effect of data below detection. In other words, the uncertainty masks the possible change.
- In all cases, the percent below MDL should be considered as a possible source of bias when interpreting site level trends.

Setting Up Data for Trend Analysis
Creating Valid Annual Averages

Data averaging is fully covered in *Preparing Data for Analysis,* Section 4, and summarized here for convenience.

- Subdaily data should first be aggregated to valid 24-hr averages. For a given day, 75% of data at the expected subdaily sampling duration is suggested for a valid 24-hr average.
- 75% of data at the expected daily sampling frequency is suggested for a valid calendar quarter average.

Frequency	75% Quarterly Completeness Cutoff
Daily	68
Every 3rd Day	23
Every 6th Day	11
Every 12th Day	5
Unassigned	5

- At least 58 days are suggested between the first and last sample in a quarter to ensure that sampling represents the entire quarter
- Data for 3 of 4 quarters are suggested for annual averages prepared from quarterly averages to ensure that sampling represents the entire year. Some air toxics concentrations show significant seasonal variations.

Setting Up Data for Trend Analysis
Creating Valid Trends

Trends are investigated for a unique combination of parameter, monitoring location, and method code.

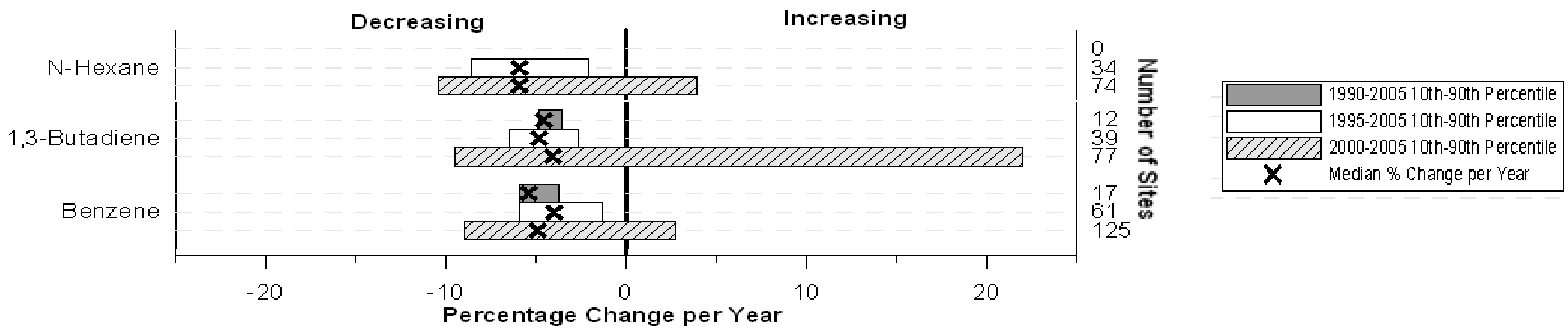
- Initially, it is important to segregate method codes for a given parameter and monitoring location to assess differences (e.g., biases, detection limits) that might result in comparability issues. In addition, methods may change over time, perhaps causing significant analytical biases that may affect trends assessments. After investigating individual trends, e.g. by method, further aggregation may be reasonable (discussed later in this section).
- At a given monitoring location, sometimes more than one monitor reports the same pollutant, known as a collocated measurement. When collocated measurements are made, data from each monitor are differentiated in AQS using POCs.

  Collocated measurements should be investigated individually as outlined in *Preparing Data for Analysis, S*ection 4. If agreement between collocated measurements is good, the data may be averaged for a given parameter, site, date, and method in order to avoid double-counting. At the national level, these data were not used.

Setting Up Data for Trend Analysis

Trend Length and Completeness

- Length and completeness criteria may be used to ensure that trends are representative of the time period of interest and that data are consistent for intercomparison among sites.
- When choosing these criteria, analysts should strive to strike a balance between maximizing available data and creating valid trends in the period of interest.
- It is easier to discern underlying trends over long time periods.
- More stringent constraints result in a reduction of available data. For example, by selecting longer trend periods, fewer sites will be available for analysis because longer continuous operation is required. On the other hand, shorter trend periods are subject to more variability, for example, because of changes in meteorology which often obscure underlying trends.



In the example, three trend periods were investigated: 1990-2005, 1995-2005, and 2000-2005. Only 17 sites in the United States collected benzene data over the 1990-2005 sampling period that met the completeness criteria. In contrast, data from 125 sites met the completeness criteria for the shorter 2000-2005 trend period. Variability for shorter trend periods is much higher.

Setting Up Data for Trend Analysis

Trend Length and Completeness

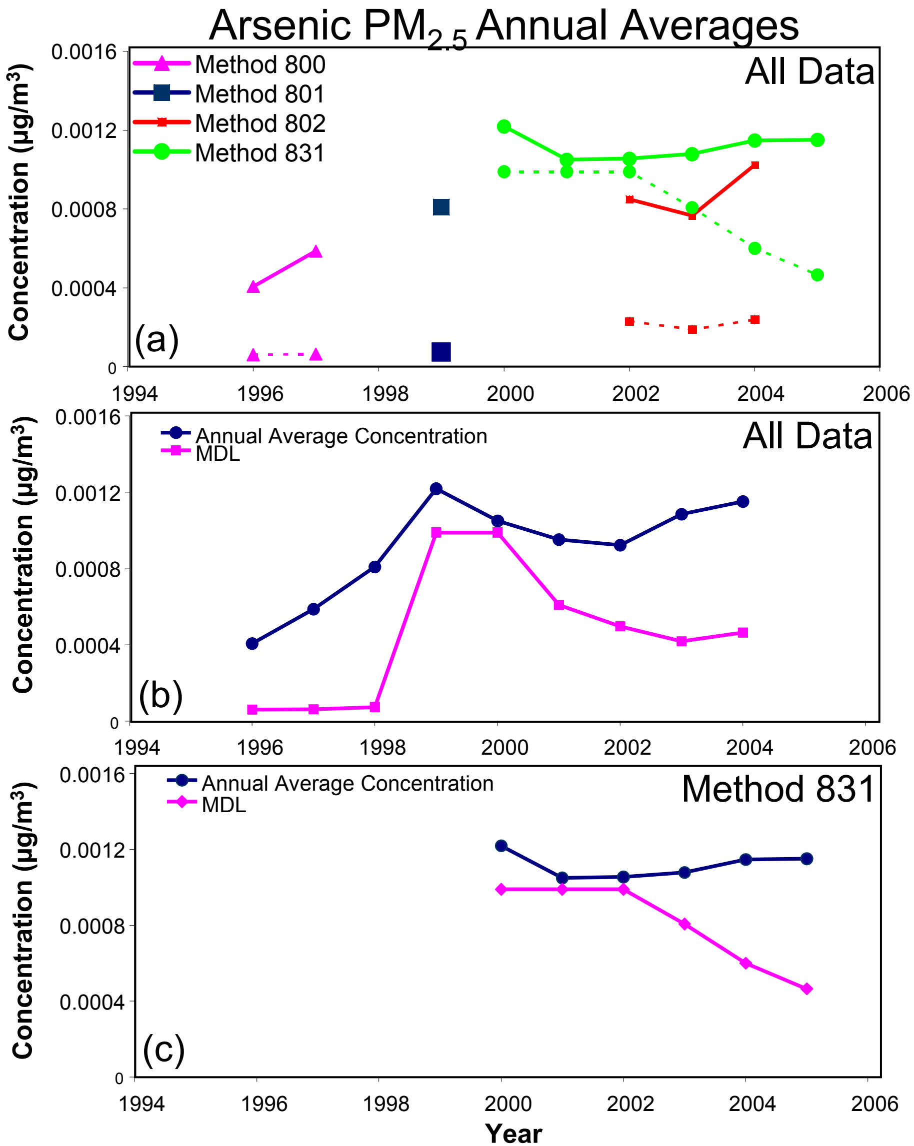
- Trend Length
  - One goal of the NATTS is to provide data with a minimum trend length of six years to be able to compare two 3-yr averages.
  - Of course, other trend periods are acceptable!
- Trend Completeness
  - Of the number of data years in a trend period, at least 75% is suggested for a site to be included (e.g., for a six-year trend period, at least five years of valid annual averages are suggested).
  - Trends with data gaps of more than two years should not be used.

Setting Up Data for Trend Analysis

Example – Creating Valid Trends

This example illustrates why looking at trends by method code is important.

- Figure (a) shows all annual averages for arsenic $PM_{2.5}$ at a site, color-coded by method. Solid lines indicate annual averages and dashed lines show average MDLs.
- Figure (b) shows the trend (blue) and average MDL (pink) for all data at a site regardless of method (i.e., the same data as in Figure (a) connected into one trend). This produces a statistically significantly increasing trend.
- Figure (c) shows the results if data are partitioned by method. Only data with method 831 are reserved because this method is the only one to have a trend period greater than four years. The results show a statistically insignificant decreasing trend, opposite the result obtained using all data.
- Which trend result is "right"?
  - The statistically significant trend in Figure (b) is driven by the lower concentration values in 1996-1998. The measured concentrations between 1996 and 2000 may be representative of ambient concentrations; however, inconsistencies in sampling method and MDLs cast doubt on the comparability of this data to post-2000 data.
  - In the end we cannot be sure which trend is "right"; more advanced analyses of the data should be undertaken if time permits. At a national level, trends could not be individually quality-controlled so they were partitioned by method to reduce inconsistencies.



Setting Up Data for Trend Analysis

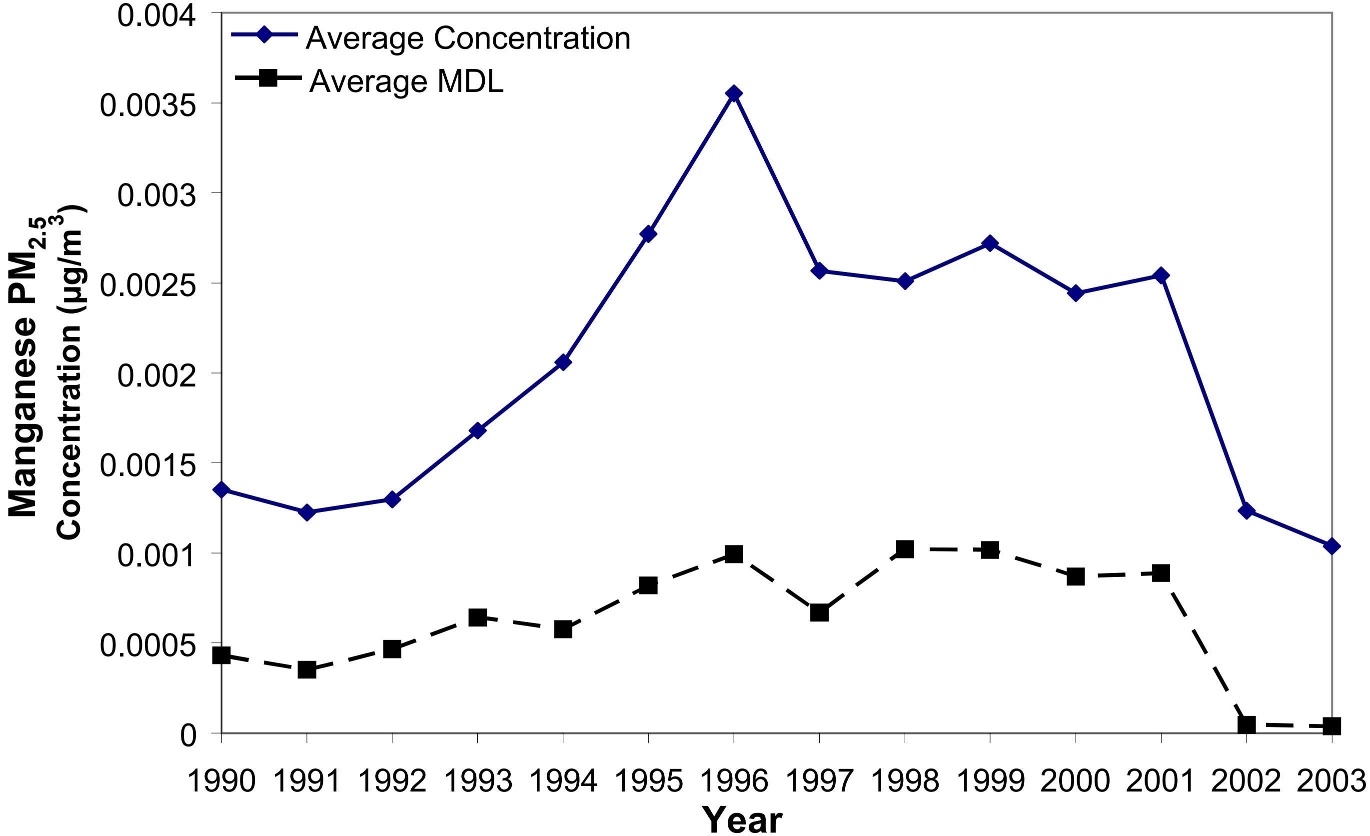
Evaluating the Effect of Method Changes

- Due to the large number of data included in the national air toxics analysis, the effect of changes in measurement methods and MDLs on trends could not be assessed on a site-by-site basis.
- During more localized analyses, such differences may be investigated; not all method changes need to be considered separately. Data may be retained across comparable method changes in order to create the longest trend periods possible.
- Assessing the comparability of methods will be a case-by-case analysis; no one procedure will provide the answer, but the following is a good start:
  - Plot all available annual averages and associated average MDLs, color-coded by method for each air toxic (as in Figure (a) on the previous slide); tabulate the percent of data below detection by year.
  - Visually assess method changes for unusual patterns in average concentration and MDL.
  - If MDL changes occur, investigate the percent of data below detection to determine if MDL/2 substitutions are driving the difference. Keep in mind the percent of data below detection and effect of MDL/2 substitutions for subsequent analyses.
  - Examine trends in air toxics data that are not expected to change significantly between years (e.g., carbon tetrachloride); significant jumps in annual average concentrations for these air toxics may indicate a problem.
  - Compare pollutants measured by the same methods that are expected to vary together (e.g., benzene and toluene) and look for discontinuities.
  - Investigate collocated data together, if available. In some cases, a measurement method may have changed in the primary monitor, but not in the secondary monitor. Look for changes in the relationship in concentrations between the monitors.

Effect of Changes in MDL on Trends Assessment

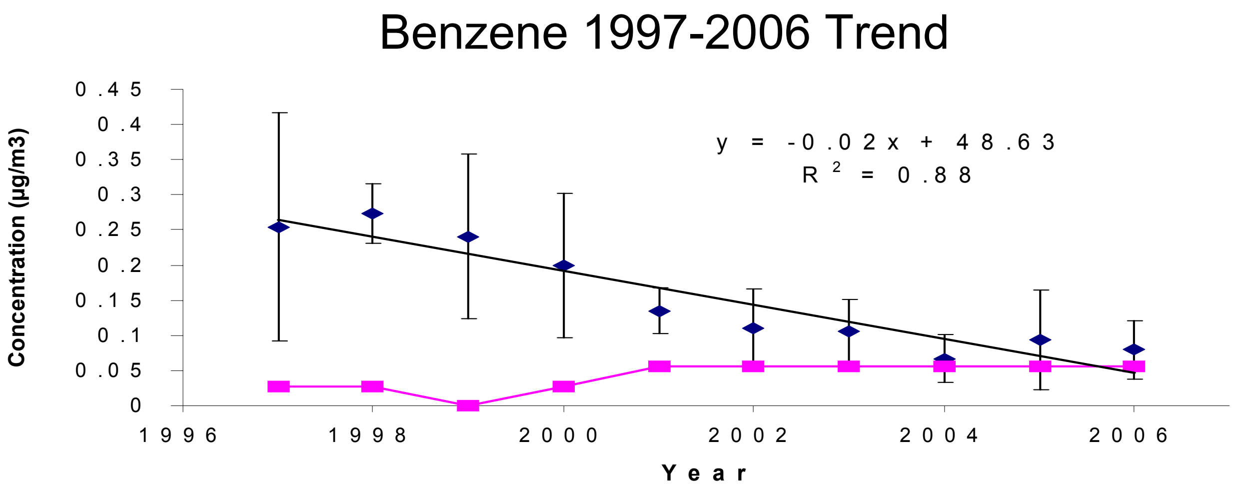
- Another important consideration in preparing data for trend analysis is that detection limits can change over time for a given monitoring site, parameter, and method. At a national scale, some detection limits change by orders of magnitude.
- These changes may influence annual averages, particularly if MDL substitutions are used. Similar trends between MDL and annual average concentrations may indicate that the changes in MDL are strongly influencing the annual average trends.
- It is recommended that the analyst inspect the trends in MDL in addition to the trends in concentration, especially for air toxics with concentrations close to the MDL (i.e., within a factor of 10).
- More sophisticated statistical analysis may be needed to quantify the underlying influence of the MDL changes on the ambient concentrations. Such analysis has not yet been performed on the national data set.

Effect of Changes in MDL on Trends Assessment *Example* (1 of 2)



In the national level investigation of manganese (Mn) trends, we noted that MDL trends were similar to concentration trends. The clear correlation between the two trend lines makes us suspicious of the reliability of the overall ambient trend. This example shows average Mn $PM_{2.5}$ concentrations and MDLs from 1990 to 2003. For this data set, Hyslop and White (2007) showed that reported MDLs are much lower than actual detection limits. Current recommendations are to be cautious with data within a factor of 6 to 10 of the reported MDL. The trend shown here may not be a real trend—these data may all be below detection.

Effect of Changes in MDL on Trends Assessment *Example* (2 of 2)



In contrast to the previous Mn $PM_{2.5}$ trend, this benzene trend does not show influence from a change in MDL (i.e., the trends in concentration and MDL show different patterns).

Quantifying Trends
Approach

- Initial investigation of trends
  - Inspect first and last year of the trend period or two multi-year averages for change.
  - Use simple linear regression to determine the magnitude of a trend over the trend period.
- Quantifying trends
  - The percent difference between the first and last year of the trend period provides a rough, first cut, sense of the change.
  - The difference between two multi-year averages provides another measure of change and helps smooth out possible influences of meteorology.
  - The percent change per year is provided by the slope of the regression line. This "normalized" value allows the analyst to compare changes across varying lengths of time (i.e., sites with different trend periods).
- Testing the significance of the observed trends
  - Calculate the significance of the slope using the F-test (see next slide). The F-test provides a statistical measure of the confidence that there is a relationship between the two variables (i.e., the regression line does not have a slope of zero which would indicate that the dependent variable is not related to the independent variable).
  - Other methods can be employed to test for significance including t-tests, nonparametric tests (tests for and estimates a trend without making distributional assumptions such as Spearman's rho test of trend; Kendall's tau test of trend), and analysis of variance.

Quantifying Trends

Interpreting Linear Regression Output

– Example output from a linear regression of annual average benzene concentrations (performed in Excel) is provided:

Slope	Intercept	% Change	% Change Per Year
-0.3943	789.562	-69.241021	-6.2946382
R^2			
0.794456			
F-Statistic		P-value	Confidence level
30.92103			99.946575

This example output shows a decline in annual average benzene concentrations over time with 95% confidence and slope not equal to zero.

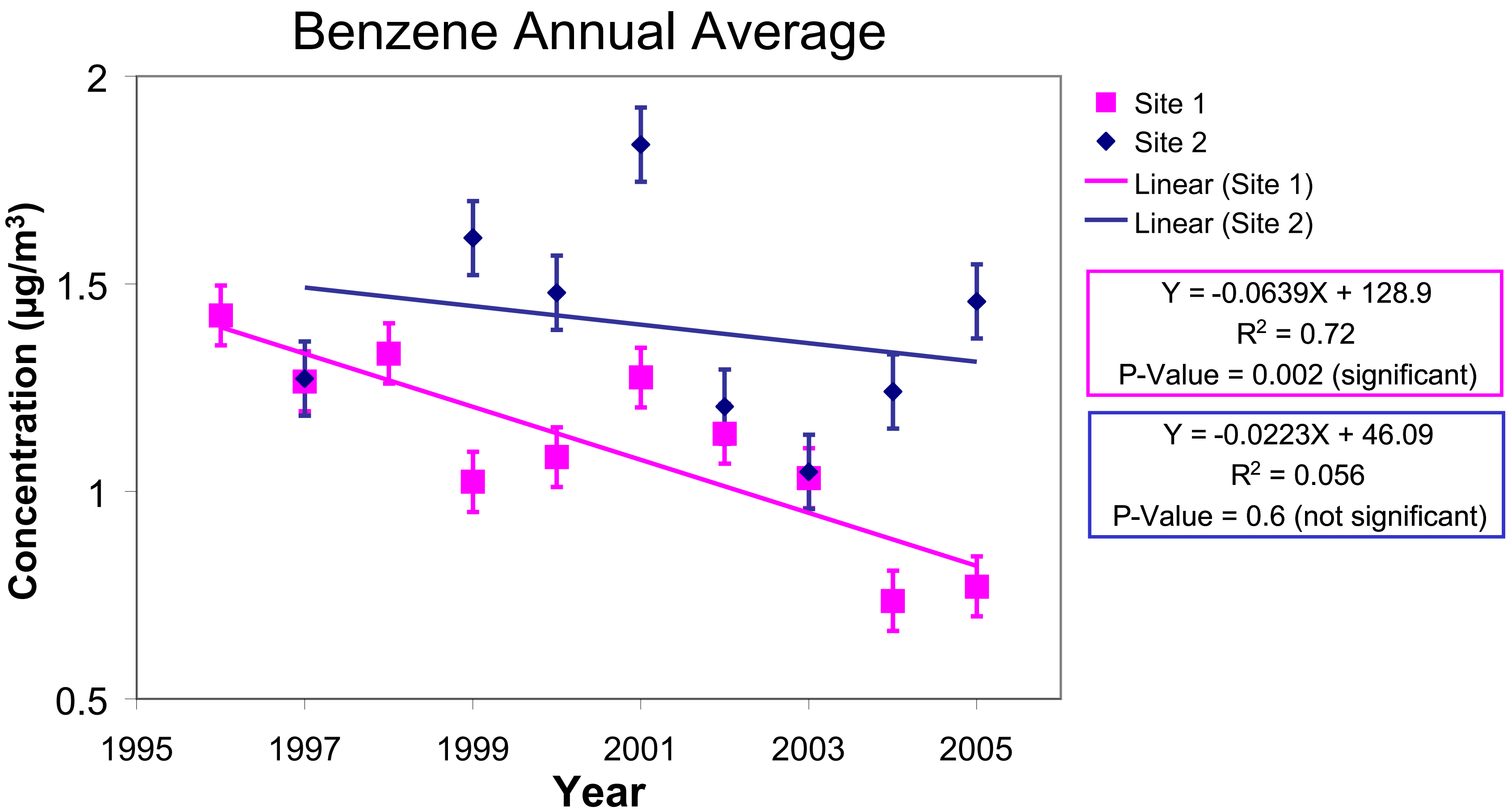
– The output is interpreted as follows:

- **Slope, intercept, % change, % change per year, R^2.** Indicate the slope of the line, y-axis intercept, % change between first and last year of the line, % change divided by number of years, and fraction of variation accounted for.
- **F-statistic or F-ratio.** F-ratio is used to test the hypothesis that the slope is 0. The F-ratio is large when the independent variable(s) helps to explain the variation in the dependent variable. Therefore, large F-ratios indicate a stronger correlation between the two variables (i.e., the slope of the regression line is NOT zero).
- **P-value.** The P-value is the probability of exceeding the F-ratio when the group means are equal (generally, 95% confidence is used as a cutoff value, corresponding to a P-value of 0.05).

– Microsoft Excel and SYSTAT11 are two of many software programs that can calculate the F-test.

Quantifying Trends

Statistical Significance Example



This example shows benzene trends at two sites. Both sites show a linear regression with a negative slope, but only Site 1 shows a statistically significant decrease. At Site 2, a decrease in concentrations is apparent, but the change is not statistically significant (i.e., failed F-test).

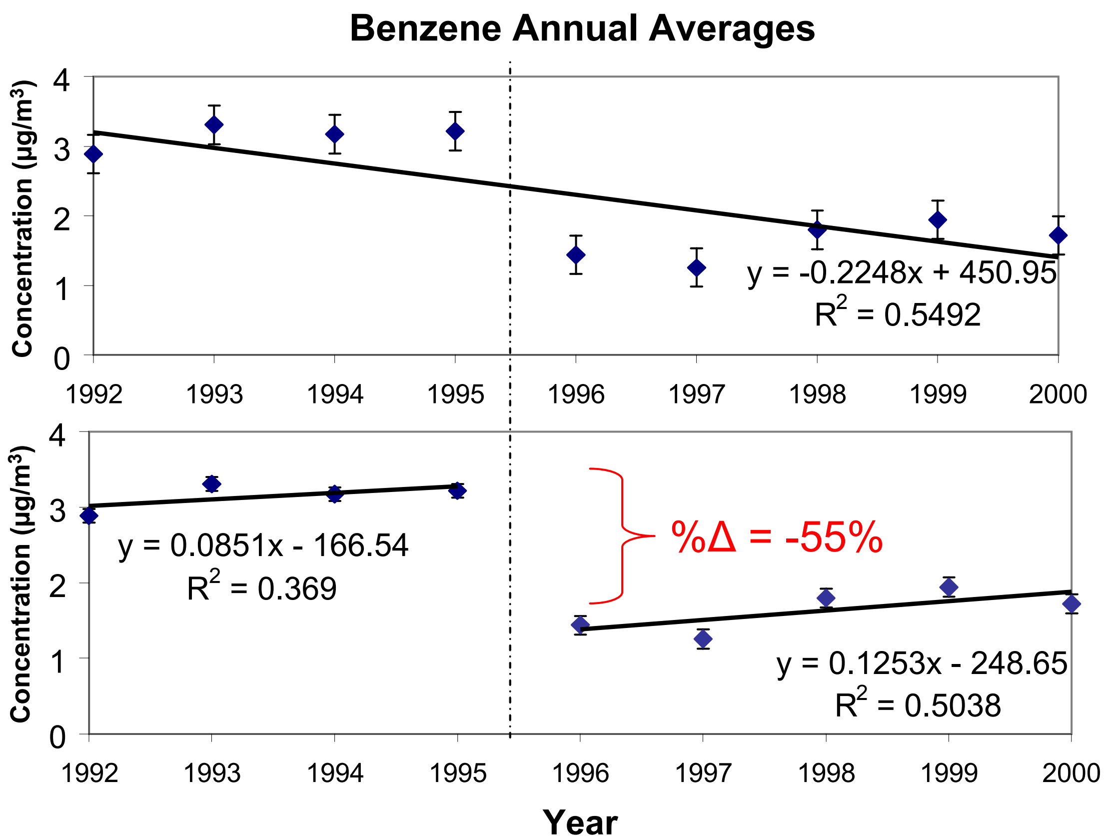
Visualizing Trends
Overview

- Visual inspection of trend data is vital! A linear fit to a trend may not be appropriate; for example, a step change may have occurred due to a major emissions regulation or a nonlinear or exponential fit may be more appropriate.
- Methods for visualizing the data include
  - Line graphs of selected indicators
  - Box plots (high and low values, median values, outliers)
  - Plots of mean or median values with confidence intervals
  - Combination of a map and temporal information

Visualizing Trends

Line Graphs

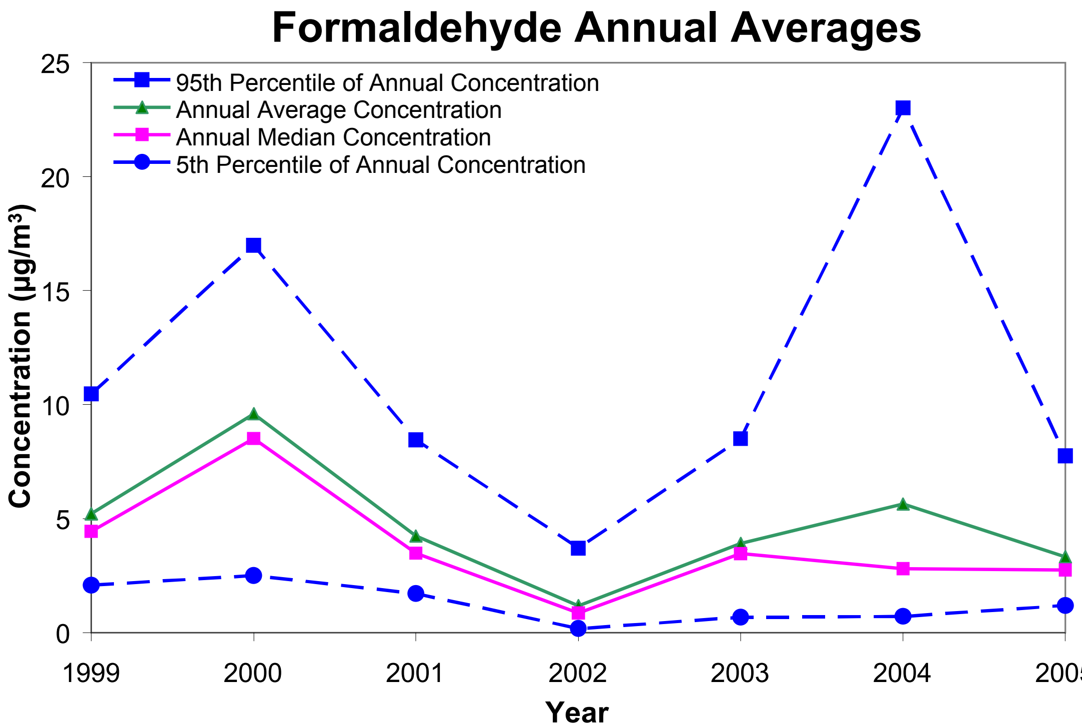
- It is sometimes useful to break a long-term trend into shorter time intervals because of significant changes in emissions. Trends should be individually and visually investigated.
- For example, benzene in gasoline was significantly reduced in several urban areas starting in the mid-1990s when reformulated gas (RFG) was introduced. Dramatic reductions were observed in ambient benzene concentrations over this time period.
- Both plots contain the same data. If one trend line is used, the overall trend decreases. If two trend lines are segregated by the RFG year (1995), the benzene concentrations are relatively flat before and after RFG implementation.
- In this case, the difference between the two time periods may be a better quantitative reflection of how benzene concentrations have changed.



The figure shows the same benzene annual averages fitted with regression lines in two ways. The first fits all data with one regression line and the second takes into account a large step change that occurred from regulations put into effect in 1995. The figure was created in Microsoft Excel.

Visualizing Trends
Using Other Statistical Metrics

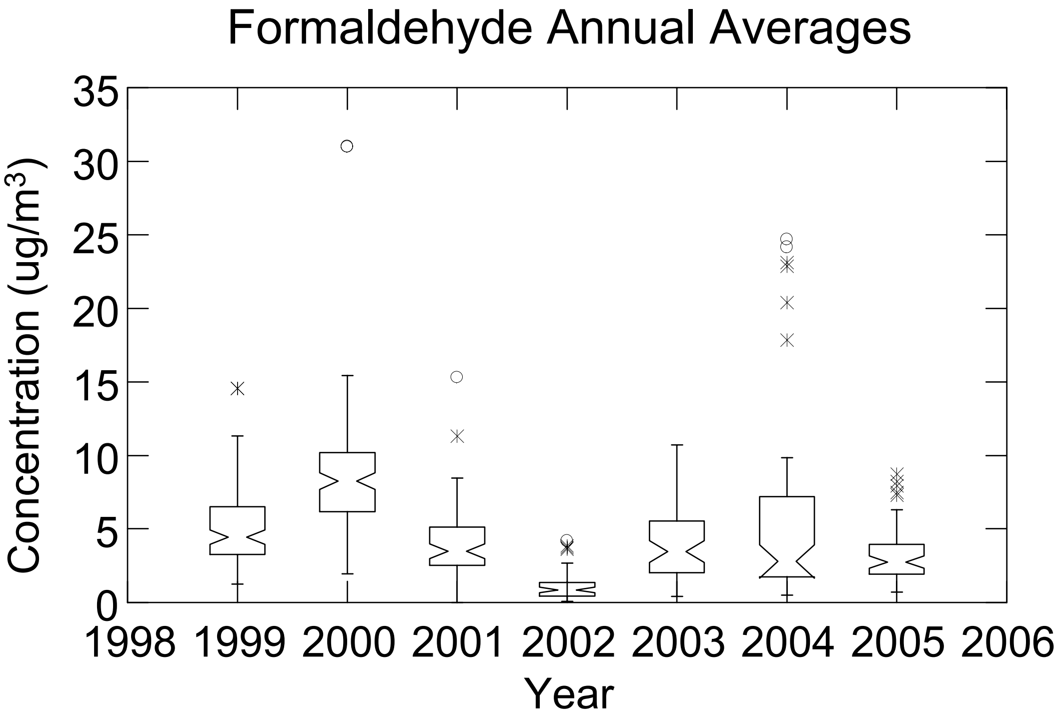
- We are typically interested in air toxics annual average trends because the annual average is used for comparisons to levels of concern for chronic health effects. Guidelines for preparing annual averages were provided previously.
- In addition to an annual average, other statistical indicators can be used to verify a trend.
  - These include median, maximum, minimum, and selected percentiles.
  - These metrics are especially helpful in identifying effects of censored data below detection.



This figure, showing formaldehyde annual data with various statistical measures, demonstrates that the annual pattern in concentration is relatively consistent. 2002 concentrations were low and there is no consistent trend over this 1999-2005 time period.

Visualizing Trends
Box Plots

- Box plots are another useful way to display multiple statistical metrics and visually asses statistical significance.
- Box plots illustrate the trends in the high and low values, interquartile ranges, median, and confidence intervals of the annual average.
- The box plots displayed here are described in *Characterizing Air Toxics* Section 5.

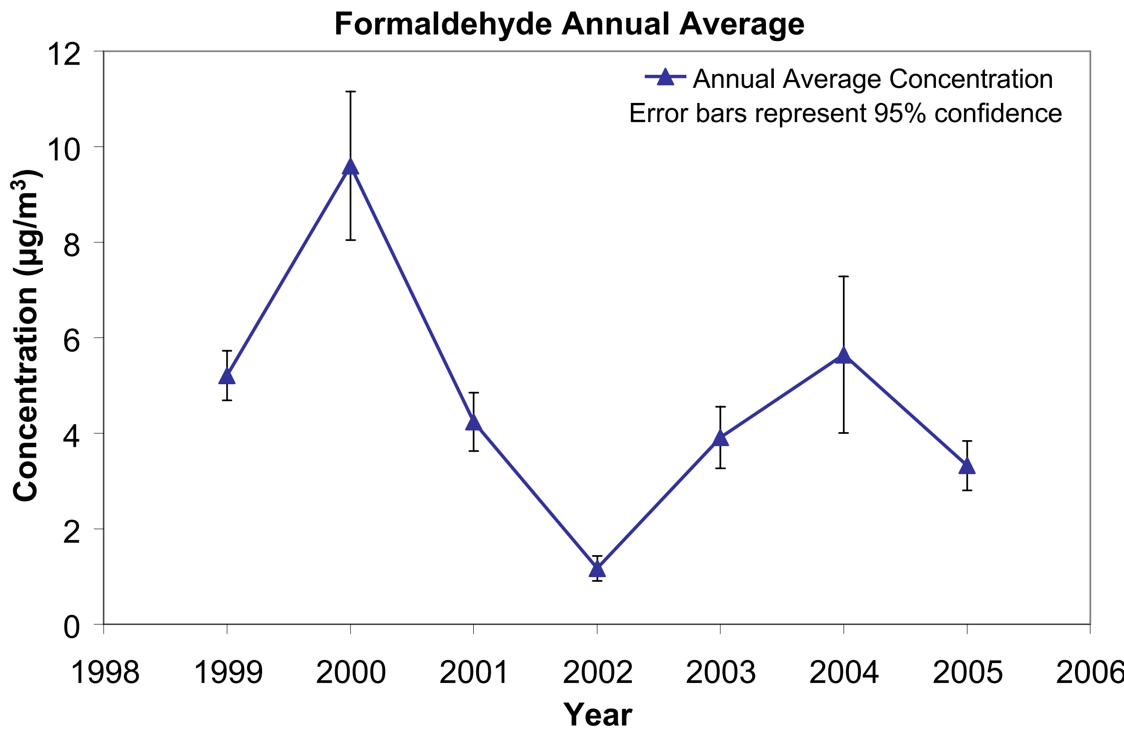


The figure shows annual formaldehyde concentrations represented as box plots. The variability is similar from year to year since the boxes for each year are about the same height. Concentrations in 2002 were statistically significantly lower than in other years because the confidence intervals do not overlap any other year.

Visualizing Trends

Using Confidence Intervals

- Confidence intervals (CIs) are shown around the annual averages for several years of data.
- Since the plotted CIs overlap in 1999 and 2001 but not in 2000 and 2001, 1999 and 2001 concentrations are not significantly different, but 2000 and 2001 concentrations are significantly different.
- CIs are a function of fewer samples resulting in large CIs. Air toxics data sets are typically small (i.e., only a few samples per month); thus, CIs help analysts understand the range in which the annual mean concentration can statistically fall.
- CI is computed as follows:



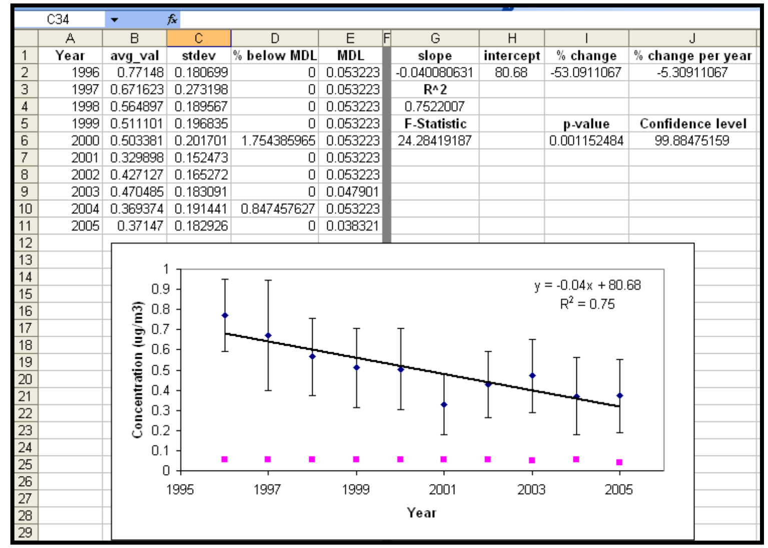
$$\overline{x} \pm z^{*} \frac{\sigma}{\sqrt{n}}$$

where x is the mean value, σ is the standard deviation, n is number of samples, and *z is the upper (1-*C*)/2 critical value (use a look up table for the % required) for the standard normal distribution.**

Visualizing Trends

Including Underlying Data

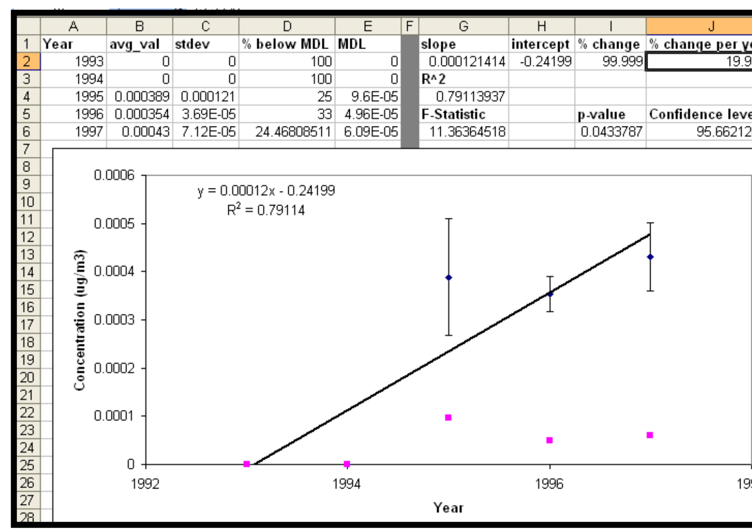
- In this example, a trend for each parameter, site, and method was plotted next to the underlying data. The figures show annual averages with standard deviations in blue and average MDLs in pink. The underlying data include the average MDL, percent below MDL by year and calculated regression, and F-value statistics as well as percent change per year.
- Figure (a) is an example of a benzene trend for the 1995-2005 trend period. In the plot, we can see that data are mostly above detection and show a statistically significant decreasing trend of about 5% per year.
- Figure (b) shows arsenic $PM_{2.5}$ data. Calculations indicate a statistically significant increasing trend of 20% per year. If these statistics were used alone, they would indicate a serious arsenic problem at this site. When the underlying data are examined though, it is clear that there may be other factors to consider. The first two years of data are 100% below detection, resulting in values that are entirely MDL/2-substituted. The values for these years may, in fact, be significantly lower and should not simply be discarded; we cannot tell from the current data. This trend should be considered suspect and validated by comparison with neighboring sites; the summary statistics should not be trusted as accurate values.



Year	avg_val	stdev	% below MDL	MDL
1996	0.77148	0.180699	0	0.053223
1997	0.671623	0.273198	0	0.053223
1998	0.564897	0.189567	0	0.053223
1999	0.511101	0.196835	0	0.053223
2000	0.503381	0.201701	1.754385965	0.053223
2001	0.329898	0.152473	0	0.053223
2002	0.427127	0.165272	0	0.053223
2003	0.470485	0.183091	0	0.047901
2004	0.369374	0.191441	0.847457627	0.053223
2005	0.37147	0.182926	0	0.038321

slope	intercept	% change	% change per year
-0.040080631	80.68	-53.0911067	-5.30911067
R^2			
0.7522007			
F-Statistic		p-value	Confidence level
24.28419187		0.001152484	99.88475159

(a)



Year	avg_val	stdev	% below MDL	MDL
1993	0	0	100	0
1994	0	0	100	0
1995	0.000389	0.000121	25	9.6E-05
1996	0.000354	3.69E-05	33	4.96E-05
1997	0.00043	7.12E-05	24.46808511	6.09E-05

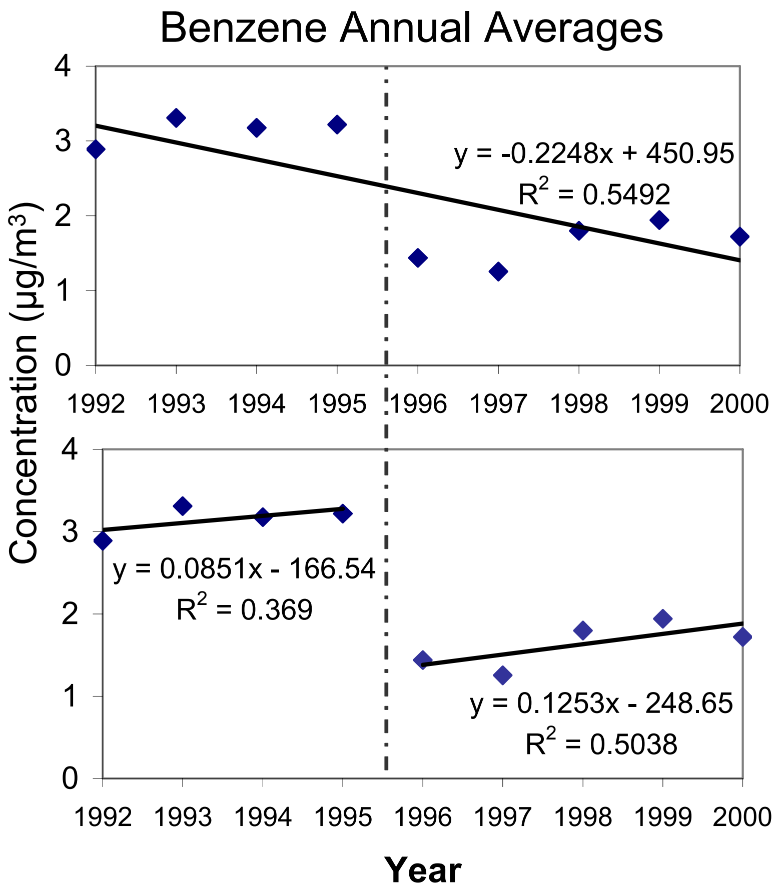
slope	intercept	% change	% change per yea
0.000121414	-0.24199	99.999	19.99
R^2			
0.79113937			
F-Statistic		p-value	Confidence level
11.36364518		0.0433787	95.662126

(b)

Visualizing Trends

Calculating Trend Period Percent Change

- There are many methods for calculating trend-period percentage change. Four such methods are listed below along with the associated percentage change that would result from applying each method to the benzene data pictured at right:
  1. Using the first and last measured data point (-40.43%).
  2. Using the regression equation (-57.12%).
  3. Using all values before and after a step change (-55.29%).
  4. Using three-year averages before and after a step change (-53.71%).
- In method 1, there is no sense of the underlying pattern for all years of interest, and the results are affected by the differences in the meteorology of the chosen years.
- Method 3 is a better measure of the percentage change because it isolates the two data points having the most impact on the overall trend, but requires visualizing the data first.
- Methods 2 and 4 use values that are weighted by more years of data within the trend period, providing more smoothing of variability from meteorological fluctuations.
- There is no right method for calculating trend results, but knowledge of possible biases of each is important when deciding which to use.



The figures show two ways to apply trend lines to benzene data from a mobile source-impacted site in California that shows a large step change between 1995 and 1996 when RFG was implemented. The figure was created with Microsoft Excel.

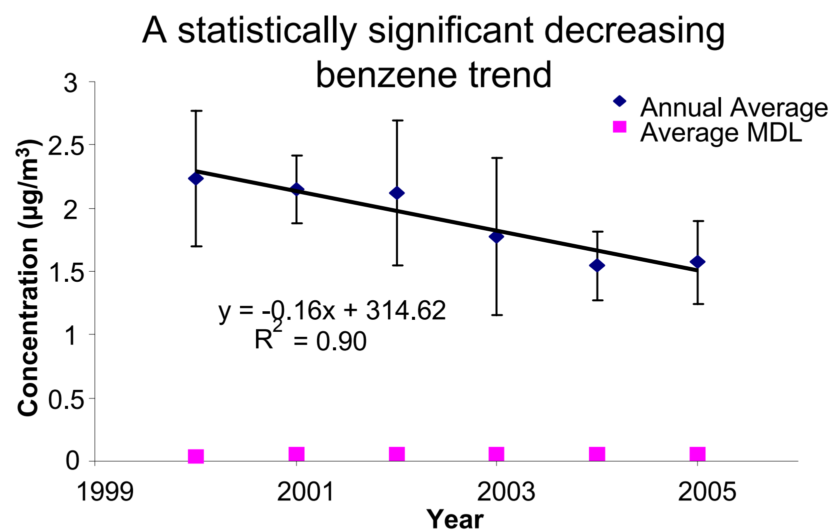
Summarizing Trends

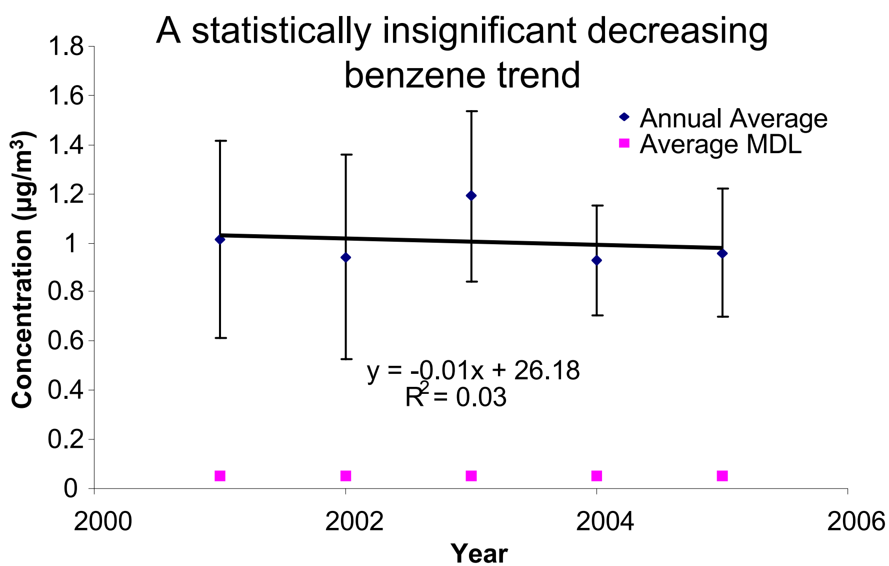
Overview

- Investigate trends among sites by pollutant.
  - Similar trends results among the sites makes a compelling argument that change on a larger spatial scale has occurred.
- Characterize the spatial distribution of trends by showing trends at each site on a map.
  - Trends may not agree nationally in direction or magnitude but may show spatial patterns of interest.
- Characterize the distribution of individual site trends by displaying the range of percentage change per year over various trend periods and for all sites meeting minimum trend criteria.

Summarizing Trends
Trends Among Sites

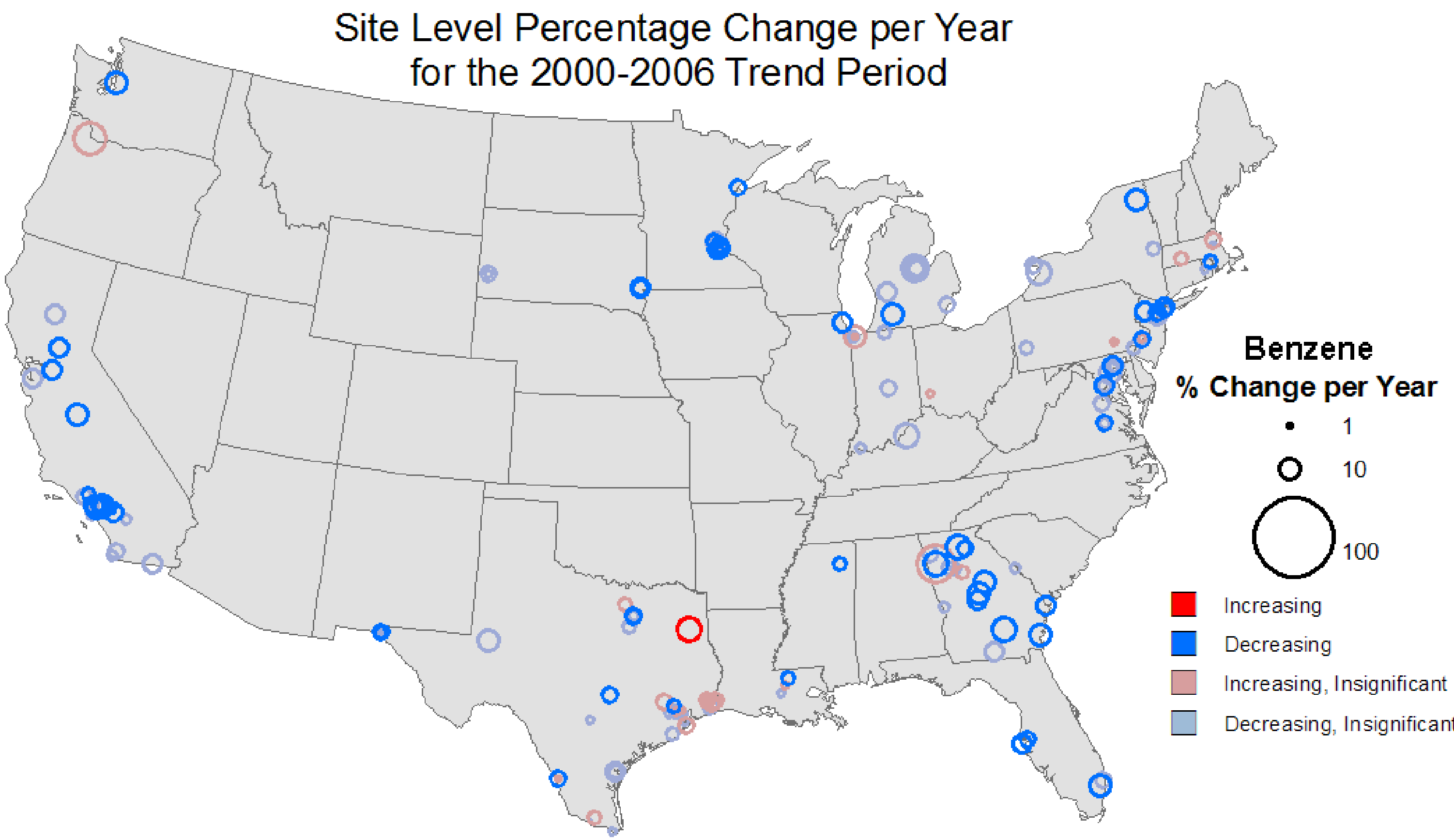
- Site-level trend investigation is vital!
- The figures show site-level trends for benzene from two U.S. sites; average MDLs are plotted in pink for reference.
- The top figure shows a statistically significant decreasing trend, while the bottom figure shows a statistically insignificant decreasing trend.
- Confidence in these results is high. The data are mostly above detection, MDLs are consistent for the whole trend period, and no outliers appear to influence the trend.
- If any of these problems do exist, the underlying trend data should be evaluated more carefully to understand the reliability of the trend.
- Next steps in investigating suspect trends
  - If one or more annual averages are an outliers, re-validate the underlying data. Is one high concentration event the cause, or is there a distribution of high values? Is there an explanation for the high annual average to prove it valid (e.g., increased local source emissions) or in error (e.g., unit conversion error)?
  - If MDL changes occur and
    - A low percentage of data is below detection, the change in MDL should not have a noticeable effect.
    - A high percentage of data are below detection, there is decreased confidence in the trend. If MDL/2 substitutions is used check that the trend does not follow the shape as the MDL changes; if it does the trend is likely unreliable.
  - If a high percentage of data is below detection without an MDL change, the central tendency of the data may still be accessible, but there is lower confidence in the trend.





Summarizing Trends

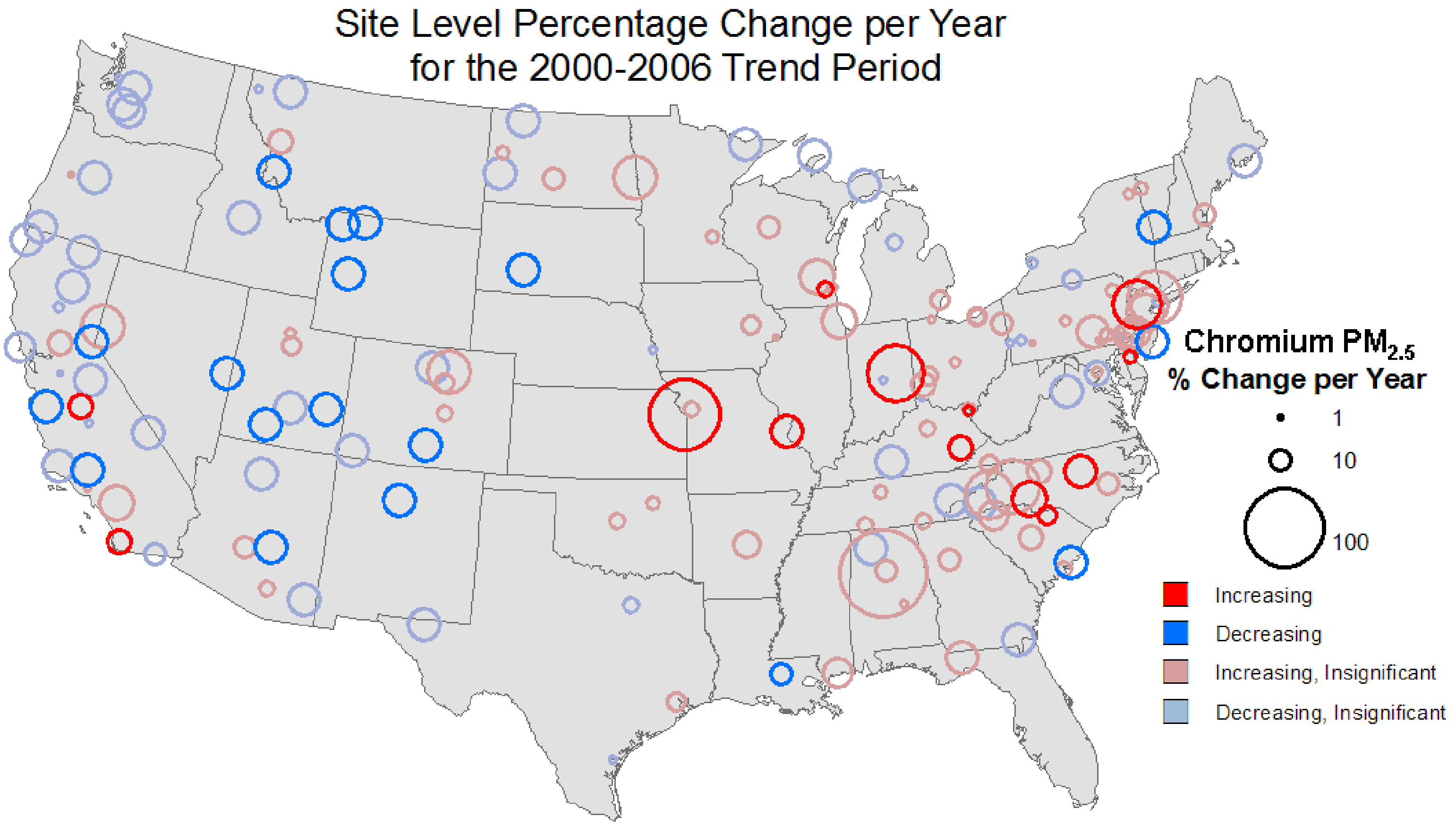
Example – Spatial Distribution (1 of 2)



This map shows the benzene site-level percentage change per year for 2000-2006. Many sites in the United States show a statistically significant decline in benzene concentrations over the period. The sites exhibiting increases over that time are typically not statistically significant trends. These data, suggest relatively high confidence that national benzene concentrations are declining nationally compared to the 2000 level. Statistical significance was quantified using the F-test at the 95% confidence level.

Summarizing Trends

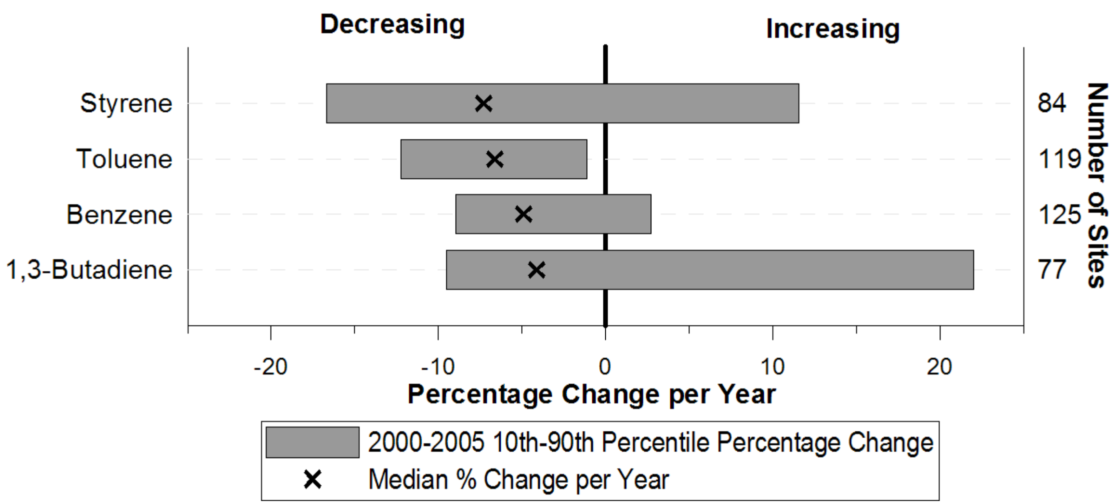
Example – Spatial Distribution *(2 of 2)*



This example shows chromium $PM_{2.5}$ concentrations across the United States in 2000 to 2006. The statistically significant trends are spatially distinct, indicating increasing concentrations in the eastern half of the country and decreasing concentrations in the West.

Summarizing Trends
Example – Percentage Change per Year

- We are typically interested in how a pollutant trend at a site compares to other sites. Summarizing the data in this way provides a succinct national perspective.
- The bar chart summarizes trends in % change per year for selected mobile source air toxics for 2000-2005 data. The 10th, 50th, and 90th percentile of site-specific percentage change per year are plotted. The number of sites included in percentile calculations is also provided.
- A range of results is seen across the network (i.e., 10th to 90th percentile sites); however, most sites are experiencing declines of a few % per year with remarkable consistency (see median); "outlier" (e.g., 95th percentile) sites may be candidates for additional investigation.
- 1,3-butadiene and styrene show a wider range of % changes by site. The median U.S. monitoring site, however, shows a trend of about -5%, in agreement with the other mobile source air toxics.
- Benzene and toluene show similar ranges in % change per year and less variability in trends across the U.S. than 1,3-butadiene and styrene.
- Toluene is decreasing at 90% of sites by about 2% to 12% per year, while benzene is decreasing at most sites and may be increasing at some sites.
- The map shows the site-specific % change values for benzene used in the bar chart, similar to the proportional maps shown previously. The magnitude of the change per year is characterized by the size of the arrow. Information as to whether the trend was statistically significant is indicated by the color of the arrow.
- Comparing data summaries, such as the bar chart, to more detailed plots, such as the map, offers an overview of the data. The map shows the spatial distribution of data included in the summary statistics. For example, benzene is increasing in some areas of the United States, but none of the trends are statistically significant. Many of the decreasing trends, on the other hand, are statistically significant.



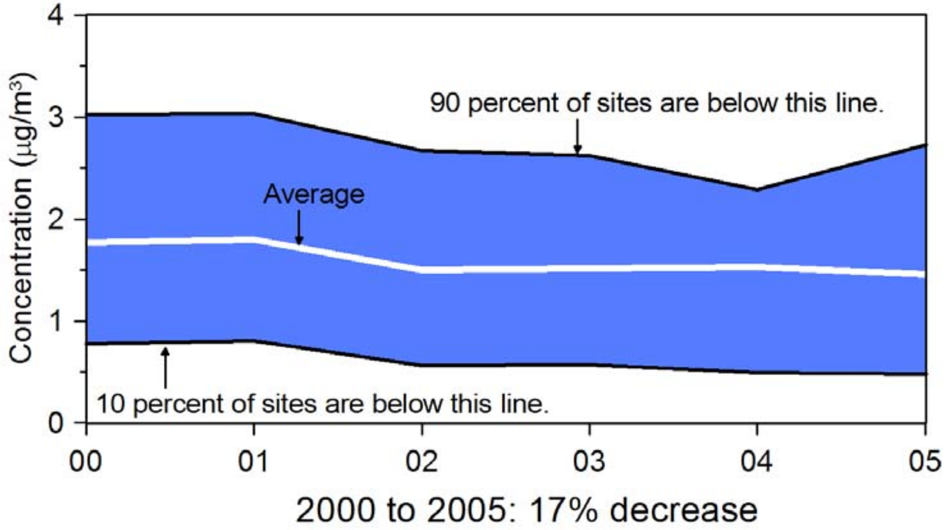
Aggregating Trends to Larger Spatial Regions

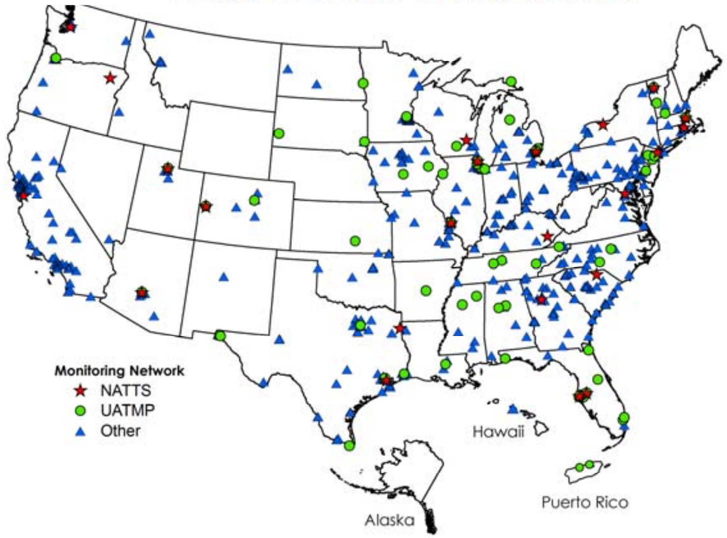
- Aggregated trends for larger spatial regions, such as trends by state or EPA Region, may be of interest to communicate results at a "big picture" level to interested stakeholders.
- Previous examples provide approaches to handling data at an aggregate level at spatial resolution less than the national scale, including summarizing percent change by year, using central tendency statistics, and plotting results on a map.
- As data sets become smaller—i.e., the analyst looks at fewer sites and fewer years—gaps in the data record become more important. For example, some site-level trend periods may meet the minimum criteria but will still have gaps in the data. Problems arise when, in combining data sets, a site, especially one measuring high or low concentrations, has missing data during some time periods.
- To handle these data gaps, the following steps are recommended.
  - For general site-level analyses, these gaps should be left as-is.
  - While not done at a national level, when aggregating to larger spatial regions, data gaps could be filled in, using the following methods, to be consistent with current trends analyses performed for criteria pollutants:
    - Missing the last year: set the missing year equal to the second-to-last year.
    - Missing the first year: set the missing year equal to the second year.
    - Missing any other year: interpolate between the adjacent two years.
    - No more than two years in succession can be missing *(this was applied in the national analyses)*.

Aggregating Trends

Example – Using Line Graphs

- Line graphs can be used to assess trends in selected indicators.
- National benzene trends (annual average concentrations) from 2000-2005 are summarized in the graph. Sites included in the summary are shown in the inset map. These types of summary displays are useful in showing general trends for multiple sites such as nationally (shown here).





Line graph figures were created with Grapher7; maps were produced in Arcmap.

Accountability

Overview (1 of 2)

- The term accountability in this section is used to refer to tying annual trends in pollutant concentrations to control programs.
- Changes in air quality may be due to a number of factors. Trends in air quality can provide evidence that local, regional, or federal emissions controls have successfully reduced ambient concentrations of pollutants harmful to human health.
- Analysis should bring as much information to bear on interpretation of trends as possible including evaluation of other potential sources of the compound in question as well as regulations, and meteorological influences that may impact emissions.
- The evaluation of the impacts of regional control programs (those that affect multiple states) and local control programs (those that affect an urban area) on air quality is complicated and is stepwise and site- and pollutant-specific.
- A major challenge in this type of analysis is the scale of influence of a control and of the impact of that control on air quality. Previous investigations of ambient air quality changes encountered the confounding influences of multiple controls applied within similar time frames and at different spatial scales.

Accountability
Overview *(2 of 2)*

- Use caution – Matching trends to changes in emissions is not sufficient to prove that an emission change actually caused the ambient change.
- Emissions regulations are typically phased in over a period of years, causing a gradual change in ambient concentrations; other factors such as meteorology, local source profiles, and MDL changes may also explain changes. The use of supplementary data (e.g., investigating trends in a pollutant <u>not</u> expected to be influenced by the emission change) is necessary to be sure observed changes are truly emissions-related.
- Two approaches to a trends accountability analysis can be taken depending on the availability of information: an emission control approach (bottom up) and an ambient data approach (top down).

Accountability
Bottom-Up Approach

- Select a control measure.
- Identify the air toxics expected to be affected and the available data, other controls that might have affected the pollutants, and other pollutants that may have been affected.
- Consider the spatial scale, or zone of influence (ZOI), of the control measure. Was the control applied at a single facility (monitor-specific or fence line), at an urban scale (MSA-wide), national scale (e.g., 49-state automobile emission rules), or global scale (e.g., Montreal protocol)?
- Determine the timing and magnitude of the changes. Was the control phased in over a period of time, applied to specific emitters? Phasing in a control makes it more difficult to discern the relationship between the ambient concentration change and the control change.
- Consider the magnitude of the expected air quality changes relative to the variability in the ambient data. If the inherent variability in the ambient data is very large, a small change in emissions may not be observable.
- Select the appropriate statistical metrics or approach for the analysis. Data treatments may help reduce the variability in the data so that trends can be observed.
- Develop hypotheses of expected changes, identify supporting evidence of changes, and investigate corroborative evidence of the changes. It is often helpful to test for changes in data sets or pollutants in which changes were not expected (i.e., check the null hypothesis).

Accountability

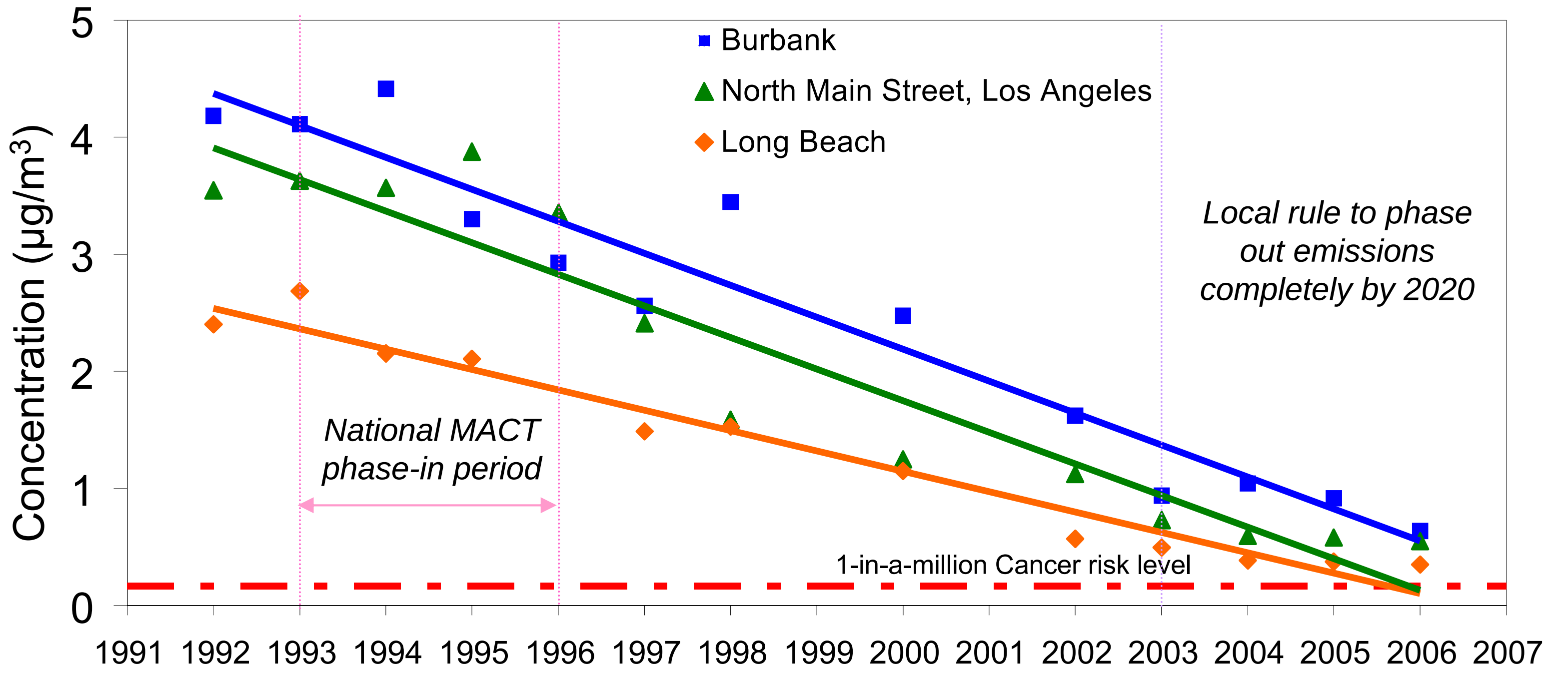
Top-Down Approach

- Quantify the change observed in the ambient data. This approach could also be applied to a pollutant for which a change was not observed but expected.
- Identify and assess other data sets and sites that may have also been affected by a similar control measure or emission change to understand the spatial scale of the ambient change. If the control was applied across a broad area, changes at additional sites might be expected.
- Identify potential emissions changes or control measures that could have contributed to the ambient trends. Local knowledge is often a key component of this part of the analysis.
- Compare the control measure implementation schedule with the ambient trends. Do the timing of the control implementation and the change in ambient concentrations coincide?
- Investigate corroborative evidence of the change and test for changes in pollutants for which a change was not expected. It is important not to over-interpret changes in ambient data.

Once methods have been developed for air toxics, it may be useful to apply meteorological adjustments to the pollutant trend. The goal is to reduce the effect of meteorology on ambient concentrations so that the underlying trend in emissions can be more readily observed. The impact of meteorology is critical when trying to assess the trend in toxics that are formed secondarily in the atmosphere (in addition to being emitted directly from sources, e.g. formaldehyde). Meteorological adjustments for air toxics have not yet been developed.

Bottom-Up Example

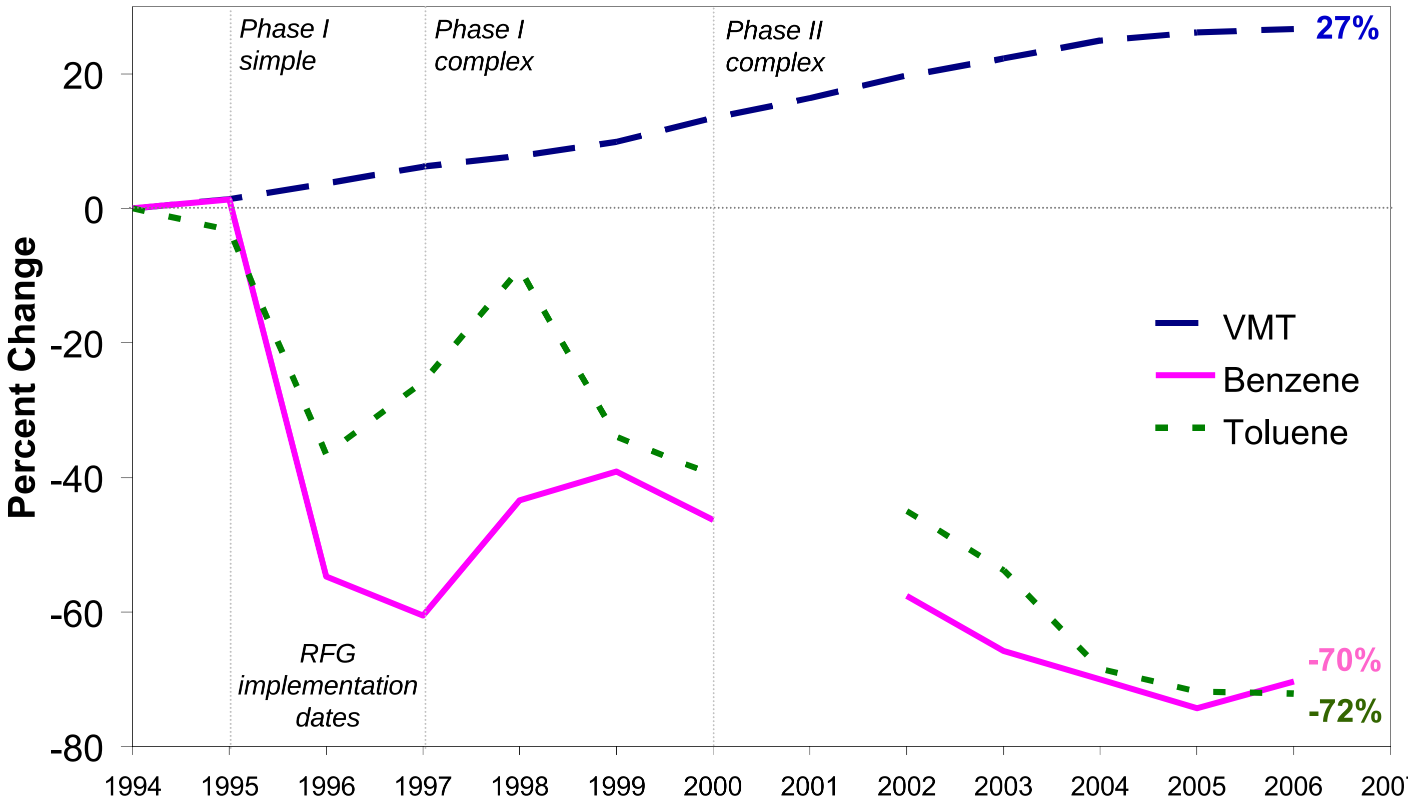
Tetrachloroethene Controls in Los Angeles



- Tetrachloroethene is the chemical most widely used by the dry cleaning industry, with over 85% of facilities using it as the primary cleaning agent. In 1993, the EPA promulgated technology-based emissions standards to control tetrachloroethene emissions from dry cleaners.
- The MACT standards implemented in 1993 resulted in drastic reductions in tetrachloroethene concentrations in the Los Angeles area where monitoring data have been available from three sites since 1992.
- Trend lines show the reductions over time in average ambient concentrations. Although concentrations in the Los Angeles area are still above the cancer risk level of concern, exposure to this air toxic has been reduced by about 80% in the past 15 years. In addition, the local South Coast Air Quality Management District implemented a rule to phase out tetrachloroethene emissions completely by 2020.

Bottom-Up Example

Ozone Precursor Controls in Baltimore, MD



- Air toxics, such as benzene and toluene, that are emitted by motor vehicles are significant contributors to ozone formation. Reformulated gasoline (RFG) was introduced in the United States in phases to reduce motor vehicle emissions of benzene and other ozone precursors in order to reduce ambient ozone concentrations.
- Benzene and toluene concentrations decreased after the 1995 implementation of RFG despite an increase in the number of vehicle miles traveled by cars and trucks in the Baltimore area.
- The largest part of the decreases in benzene and toluene concentrations is directly attributable to the implementation of RFG; the more steady, few percent change per year observed in latter years is likely due to fleet turnover (i.e., newer cars with lower emissions replacing older, more polluting vehicles).

National Level Top-Down Example
Method

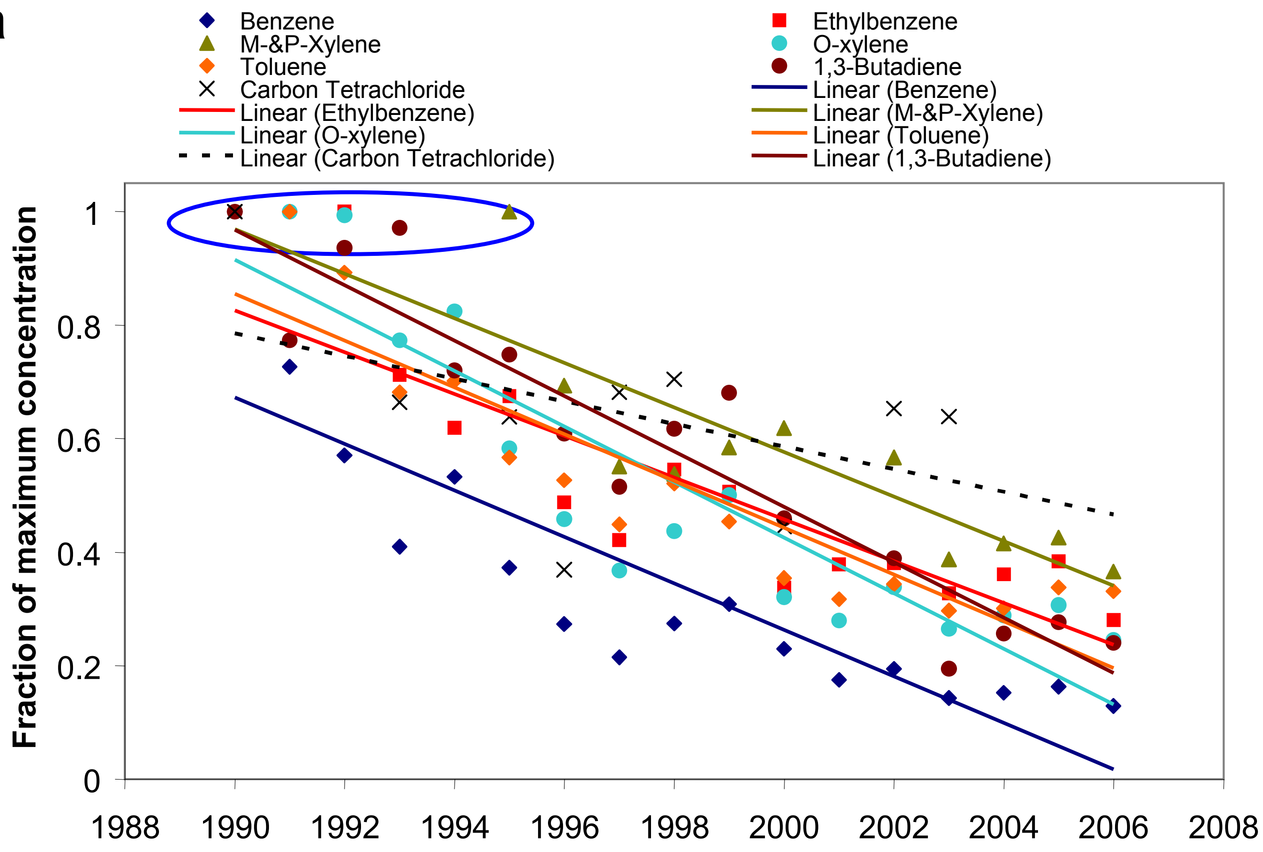
- The hypothesis is that if pollutants are emitted by the same source, emissions should covary over long time scales. In other words, trends should be parallel if normalized.
- At a national level, the goal was to identify covariant trends in MSATs as an indicator of sites dominated by mobile source emissions.
- Site-specific trends for six MSATs (benzene, 1,3-butadiene, toluene, ethylbenzene, o-xylene, m-&p-xylenes) were investigated using carbon tetrachloride as a control.
- Trends were normalized by the maximum annual average concentration within the trend period by site and pollutant (i.e., annual average concentrations each year were divided by the highest annual average in the time period for each pollutant and at each site). Normalization creates a data set that is easier to compare across sites and pollutants and shows the relative change in concentration.
- Linear regression was used to create trend lines for each pollutant.
- The sites were visually grouped into various categories by the behavior of pollutant trends. For example, if all MSAT trends had a similar slope, we expect the change in concentration at that site to be a consequence of mobile source reductions. If one MSAT exhibited a very different slope than the others, we would conclude that another source of that pollutant impacting the site was likely.
- For this analysis, only the site and parameter were required to be consistent over the trend period (method and POC were allowed to float between years). Sites with more than five annual averages were included.
- Sites were then investigated using Google Earth to see if our hypotheses were correct.

National Level Top-Down Example

Output

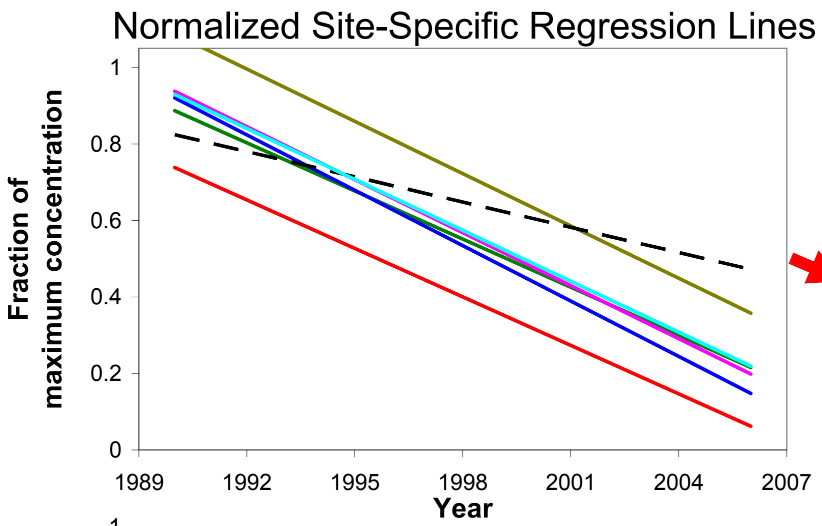
Example output from a site illustrates results of this analysis

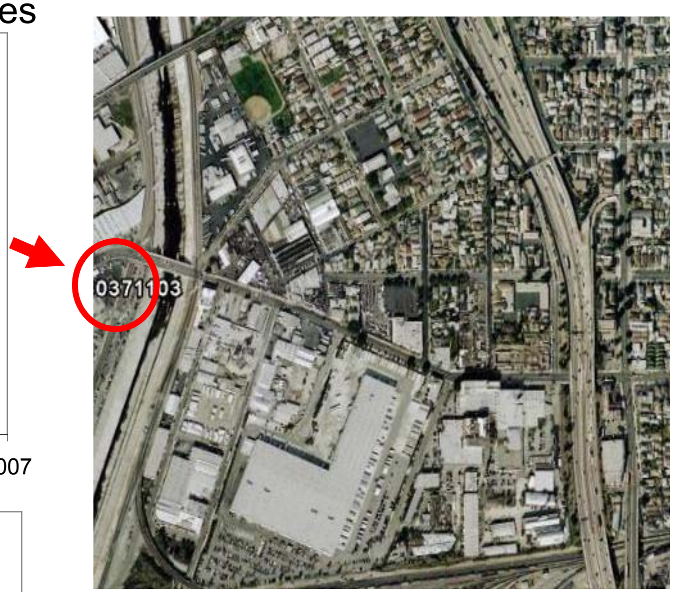
- Due to normalization, maximum values are always = 1.
- The slopes of the MSATs are close to parallel.
- Carbon tetrachloride's slope (dashed line) is very different (flatter) than the MSATs.

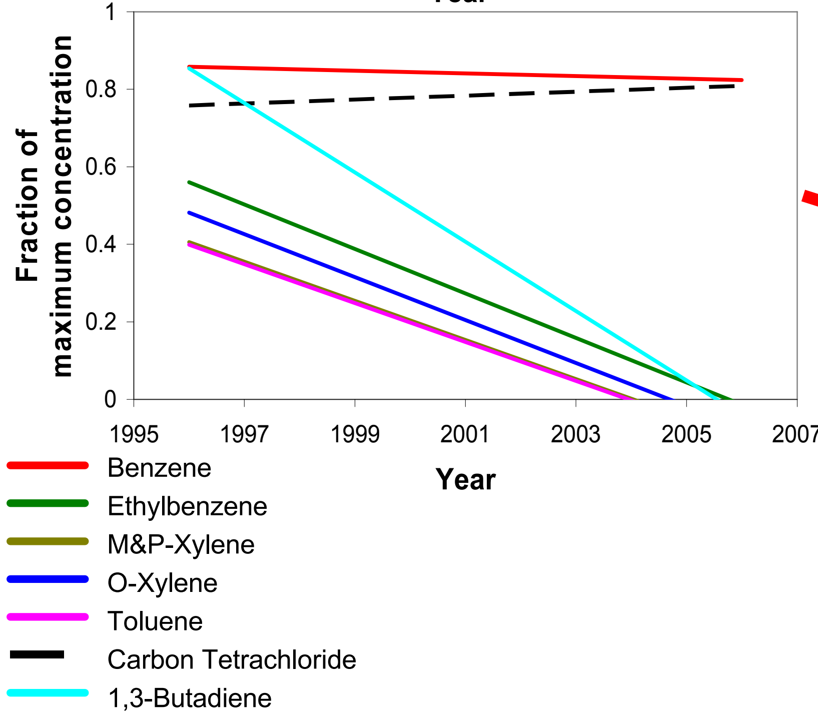


National Level Top-Down Example

- At the monitor in the top example, all MSATs show a similar declining slope. Investigation of the monitoring location indicates that this site is primarily mobile source-dominated (it is located very near a major freeway).
- The second example shows similar slopes for all MSATs except 1,3-butadiene and benzene. Benzene shows a much slower decline in concentration than the other MSATs while 1,3-butadiene shows a slightly faster decline. This monitor is located near a large refinery with both benzene and 1,3-butadiene emissions which may explain this divergent behavior.



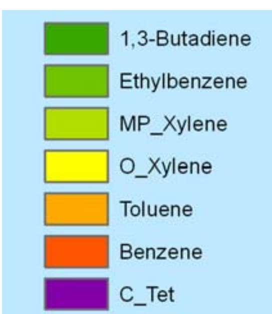


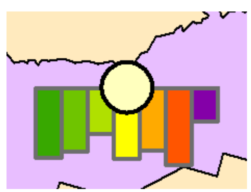


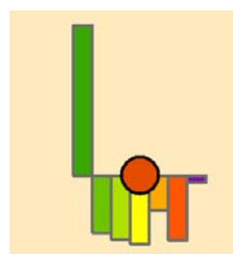
National Level Top-Down Example

Spatial Characterization of Trend Profile "Signatures"

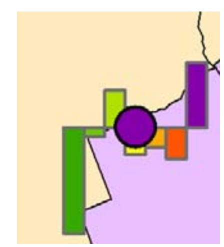
- Visual inspection of the slopes of trends provides useful information on the covariance of pollutant concentrations over time.
- The percentage change in concentrations per year can also be plotted on maps for each pollutant shown in the scatter plots to spatially investigate the trends profiles.
- Mobile source signatures have MSAT profiles of similar magnitudes; other signatures have increasing or varying magnitudes among the pollutants.




Mobile source


1,3-Butadiene

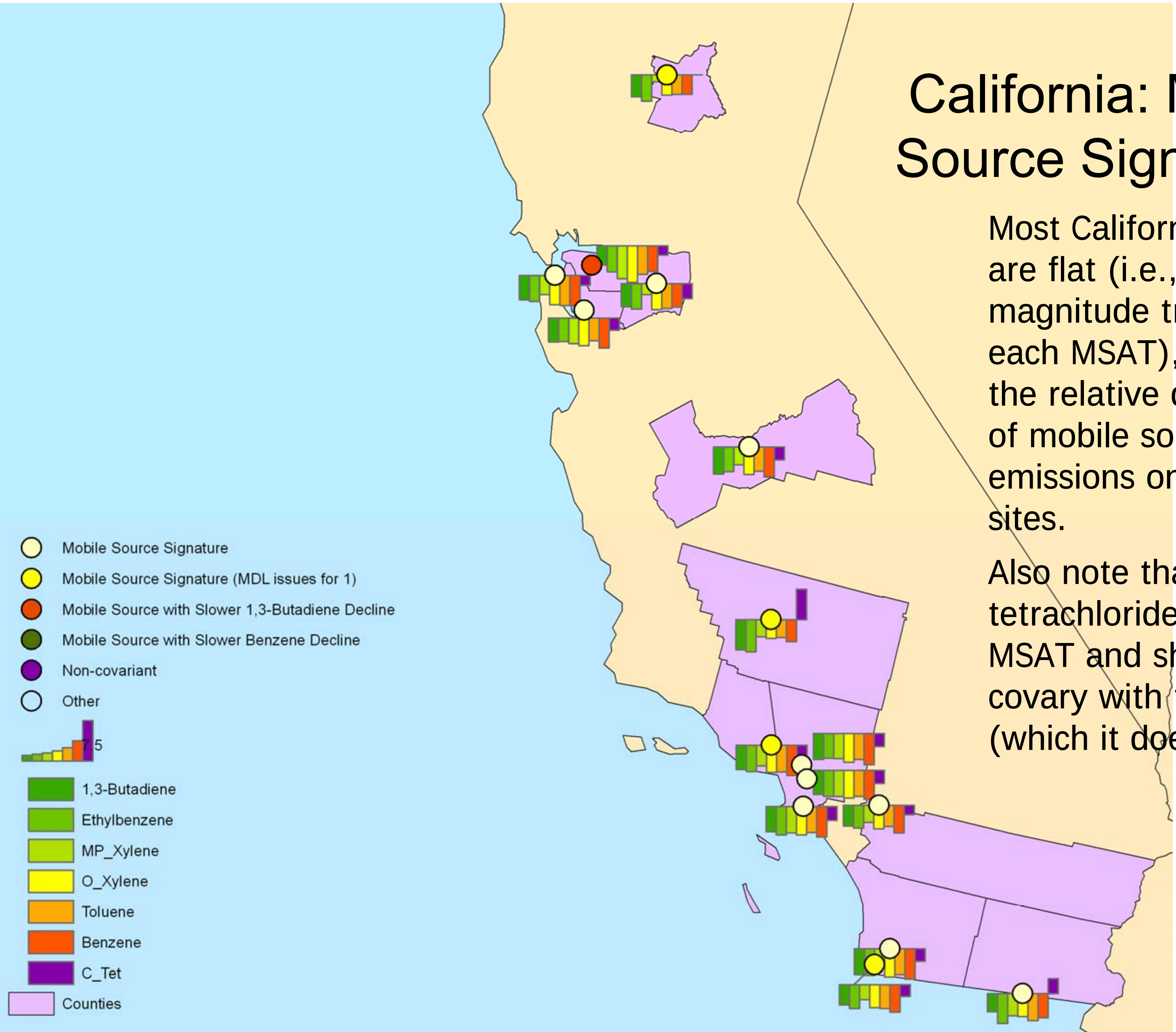

Benzene


Noncovariant

California: Mobile Source Signatures

Most California profiles are flat (i.e., similar magnitude trend for each MSAT), indicating the relative dominance of mobile source emissions on these sites.

Also note that carbon tetrachloride is not an MSAT and should not covary with the others (which it does not).



National Level Top-Down Example
Summary

- The top-down approach is a useful way to investigate site-level trends of pollutants commonly emitted by the same source.
- Most sites in the United States conformed to our expected mobile source trend profile.
- The technique also allows identification of sites at which trends do not conform to expectations. For example, two mobile source-like signatures were identified at most of the remaining sites
  - 1,3-butadiene signature sites showed shallow or increasing 1,3-butadiene (possible measurement issues?).
  - Benzene signature sites showed shallow or increasing benzene (likely explained by nearby point-source emissions for some sites but was not clear for others).
- Some sites showed increasing trends or noncovariant trends in multiple MSATs. Nearby emissions sources may be influencing trends at these sites, and they may be good candidates for case study analyses of other emissions sources.
- The top-down approach may be applicable to other pollutants from mobile sources (CO, NO_x, black carbon) or other emissions sources of multiple co-emitted pollutants.

Meteorological Adjustment of Air Toxics
Introductory Thoughts

- Meteorology can impact air quality.
  - Meteorology can vary significantly among years (e.g., El Niño), and meteorology can have a considerable effect on air quality.
  - To understand changes in air quality that are attributed to emission controls, we need to be able to adjust the data to account for meteorological conditions that were very different from average conditions.
  - By properly accounting for the portion of the variability in the data attributable to changes in meteorology, we can compare air quality among years with widely different meteorological conditions.
  - This assessment is important because we do not have control over meteorological changes.
- Using meteorological adjustment of air toxics is still being explored.
- Application of meteorological adjustment is likely at site-level, and each site and pollutant will need to be treated discretely.
- In preliminary investigations, meteorology accounted for 15-25% of total variability for benzene and lead (tsp) at selected sites; meteorological adjustments smoothed trends; and meteorological trends adjustment appeared to be important for interpretation of trends in benzene and lead (tsp) and may be important to other air toxics as well. More investigation is needed to finalize an approach for meteorological adjustment.

Resources

Tools Available for Trend Analysis

- Examples in this section were created with
  - ArcInfo and ArcView <http://www.esri.com/>
  - SYSTAT
  - Grapher
  - Microsoft Excel
- Air toxics guidance
  - http://www.epa.gov/ttn/fera/risk_atra_main.html
- Computing 95% upper confidence limit (95% UCL) for use in risk assessment
  - ProUCl 4.0 available at http://www.epa.gov/nerlesd1/tsc/software.htm

Trends Summary (1 of 2)

- Setting up data for trends analysis.
  - Acquire and validate data. See *Preparing Data for Analysis*, Section 4, for a complete discussion.
  - Identify censored data. Separate data at or below detection for each parameter, site and method.
    - Count the number of occurrences by value. Do the values indicate a specific substitution method?
    - Make scatter plots of data below detection vs. the detection limit for each value. The slope of the line will indicate the denominator if MDL/x substitutions were used, even if alternate MDLs are available.
  - Treat data below detection.
    - If uncensored values are used, include them “as is”.
    - If censored values are used, substitute MDL/2 or use a more sophisticated method as appropriate.
    - If a mixture of censored and uncensored data is used, compare the methods of all substituted vs. only censored substituted to see if results agree. If not, more advanced methods to treat data below detection may be necessary.
  - Calculate valid annual averages. See *Preparing Data for Analysis*, Section 4, for a complete discussion.
  - Create valid trends.
    - Segregate trends by parameter, site and method.
    - Consider and apply trend completeness criteria depending on data needs.
      - Minimum trend length of 6 years
      - 75% yearly completeness within trend period
      - Data gaps longer than 2 years not allowed
    - Consider yearly aggregated percent of data below detection.
      - Look at all data regardless of percent below detection
      - Remove trends where more than half the year’s data are less than 15% of data above detection

Trends Summary (2 of 2)

- Quantifying Trends
  - Magnitude of change
    - Use simple linear regression to calculate first and last year values to determine the percent change over the trend period.
    - Calculate percent change per year for intercomparison of trend periods.
  - Significance of change
    - Quantify the statistical significance of the slope using the F-test.
    - Typically, a trend is considered significant at or above the 95% confidence level.
  - Visualize trends; always include annual percent below detection as a measure of uncertainty.
    - Line graphs
    - Box plots
    - Spatial representations
  - Summarize trends
    - Characterize the distribution of percentage change per year for all sites and investigate mean, median and percentiles.
    - Characterize the spatial distribution of the percentage change per year.
    - Look for consensus in results among methods.
- Accountability – tie annual trends to control programs
  - Acquire background information on control programs; compare this information to site-level metadata keeping in mind local sources, site location etc.
    - Implementation date or time period
    - Pollutants affected and expected magnitude of reduction
    - Types of sources affected
  - Acquire emissions inventory data
    - Toxics release inventory data (TRI) (does not include mobile source emissions!)
    - National emissions inventory data (NEI)
  - Compare ambient data to emission inventories and control programs—correlation is not enough to prove causation
    - Compare similar pollutants that should experience concentration reductions resulting from the control programs.
    - Compare similar pollutants that should NOT experience concentration reductions for the control program.

Additional Reading (1 of 2)
Meteorological Adjustment Techniques

Methods for adjusting pollutant concentrations to account for meteorology

- Expected peak-day concentration (California Air Resources Board, 1993)
- Native variability (California Air Resources Board, 1993)
- Filtering techniques (e.g., Rao and Zurbenko, 1994)
- Probability distribution technique (Cox and Chu, 1998)
- Classification and Regression Tree (CART) analysis (e.g., Stoeckenius, 1990)
- Linear regression (e.g., Davidson, 1993)
- Nonlinear regression (e.g., Bloomfield et al., 1996)

Additional Reading (2 of 2)

Meteorological Adjustment Techniques for Ozone and Particulate Matter

- PAMS ozone adjustment techniques, http://www.epa.gov/air/oaqps/pams/analysis/trends/txtsac.html #meteorological
- Thompson M.L., Reynolds J., Lawrence H.C., Guttorp P., and Sampson P.D. (2001) A review of statistical methods for the meteorological adjustment of tropospheric ozone. *Atmos. Environ.* **35**, 617-630. Available on the Internet at www.nrcse.washington.edu/pdf/trs26_ozone.pdf
- Data Quality Objectives for the Trends Component of the PM Speciation Network (includes meteorological adjustment techniques in Appendix), http://earth1.epa.gov/ttn/amtic/files/ambient/pm25/spec/dqo3.pdf

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