



pennsylvania
DEPARTMENT OF ENVIRONMENTAL
PROTECTION

Bureau of Air Quality

*Analysis of Long-Term Particulate Matter Monitoring Data and Toxic
Metals Health-Based Screening of Ambient Air in Bath Borough,
Northampton County, PA*

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I. Introduction

In 1995, the Pennsylvania Department of Environmental Protection's ("DEP") Northeast Regional Office ("NERO") Air Quality program staff initiated continuous, 24-hr ambient air sampling for total suspended particulate ("TSP"), sulfate and select metals at a location near the Keystone Cement Company cement production facility in East Allen Township, Northampton County, AIMS ID 20-2268581-1 (Keystone Cement facility). The community has a population of approximately 2700 residents¹. The Keystone Cement facility is located adjacent to the Borough of Bath, Pa.

Sampling was initiated to monitor the facility's particulate emissions in the ambient air and to evaluate the concentrations of total suspended particulate (TSP) matter in the community to evaluate potential health effects. In 2014, a second phase of sampling commenced to perform a toxic metals health effects screening of the ambient air monitored at the site.

This report summarizes both the historical surveillance monitoring (Phase I – 1995 through 2013) and the toxic metals screening (Phase II – 2014 through 2015). The analysis was initiated by DEP to help determine if continued surveillance and ambient monitoring for particulate matter is warranted.

General Conclusions of Analyses

- The samplers were sufficiently located and operated to collect near-continuous daily ambient air TSP, sulfate and iron concentrations from 1995 through 2013.
- Available meteorological data (wind speed and direction) used to site the monitors at the Keystone Cement facility demonstrated that the samplers were in a portion of a predominant downwind vector from the facility.
- TSP concentrations at the site dropped by 28% averaged over the three years following a major conversion at Keystone Cement facility from a wet to dry Portland cement method in 2009. Concentrations of sulfate and iron in the ambient air dropped by 26% and 11%, respectively, over the same periods.
- No exceedances of applicable National Ambient Air Quality Standards ("NAAQS") for particulate matter were observed during either phase of sampling. During Phase I sampling, 13 values of sulfate were observed over a California Air Resources Board ("CARB") sulfate ambient air quality standard of 25 $\mu\text{g}/\text{m}^3$ (micrograms per cubic meter); however, given the CARB standard is for particulate matter less than 10 microns (PM_{10}) fraction (not TSP as measured at 32K), it is unlikely that health was adversely affected, as a significant portion of TSP is non-respirable.
- In Phase II, 24-hour ambient concentrations of arsenic, cadmium, manganese, and lead were representatively sampled every sixth day in 2014 and 2015 and suitable for health-based toxic metals chronic exposure screening.
- Phase II data availability was less than optimal for Phase II, although by using the 95% Upper Confidence Limit ("95UCL") of the estimated mean concentration of a pollutant as a health-based chronic screening concentration, more reliable and conservative (i.e., more protective) estimates for screening are obtained.

¹ 2,963 residents recorded in 2010 U.S. Census.

- The final screening concentrations of arsenic, cadmium, manganese and lead were below health-based lifetime screening values, thus indicating that the ambient air in the monitored location does not pose a health issue due to chronic exposure to these metals. Measurable concentrations of total chromium and nickel were found only in 4% and 1% of the samples, respectively.
- For Phase II sampling, all ambient air toxic metal concentration screening values, using the 95UCL of the two-year screening period mean concentration, were below health-based chronic inhalation hazard and lifetime cancer risk values. For that reason, the observed ambient air concentrations at the monitored location would likely not pose a significant chronic health risk from those metals.

II. Background on Keystone Cement and Sampling Site

A. Keystone Cement Company

The Keystone Cement Company (“Keystone”) is a large manufacturer of Portland cement (NAICS Code 327310 – Cement Manufacturing). The Keystone Cement facility has been producing cement in the Lehigh Valley since it was founded in 1928. The company is owned by Giant Cement Holding, Inc. (a company of the Spanish firm Cementos Portland Valderrivas Group), which in turn is controlled by the international construction company Fomento de Construcciones y Contratos S.A. (FCC). Headquartered in Barcelona, Spain, FCC is a \$12.4 billion multinational construction and engineering firm. As part of FCC, Giant Cement Holding, Inc. (Giant) controls FCC’s North American aggregate production², cement manufacturing and waste disposal operations³. Keystone is one of three aggregate/cement/waste disposal companies owned and operated by Giant in the United States. This facility is unique in its use of hazardous waste as fuel. In addition, the facility has been subject to three major MACT standards.

The Keystone Cement facility is a permitted major source under Title V of the Federal Clean Air Act (TV Permit 48-00003). It is subject to the 40 CFR Part 63 National Emissions Standards for Hazardous Air Pollutants (NESHAP) for Portland Cement Manufacturing (Subpart LLL), Hazardous Waste Combustors (Subpart EEE), and Industrial/Commercial/Institutional Boilers (Subpart DDDDD) in addition to general 40 CFR Part 63 Subpart A requirements.

The Keystone Cement facility is also subject to 40 CFR Part 60 New Source Performance Standards (NSPS) for Portland Cement (Subpart F), Coal Preparation (Subpart Y) and Non-Metallic Mineral Processing Plants (Subpart OOO) in addition to general 40 CFR 60 Subpart A requirements.

² The Keystone Cement facility is collocated with a large limestone quarry providing ideal feedstock for cement clinker manufacture. Giant also sells graded aggregate as a separate product.

³ The Keystone Cement facility was the first cement plant in the U.S. permitted to use listed hazardous wastes as supplemental fuel for the rotary cement kilns. Since 1995, anywhere from 5% to 50% of the annual heat input to the kilns has been from these liquid wastes (usually solvent mixtures and/or waste oils). Bituminous coal is used for the majority of the balance heat input.

General information on the process of making Portland cement and the emissions associated with cement manufacturing can be found in the EPA's AP-42 compendium of emissions factors.⁴

Prior to 2009, the Keystone Cement facility employed a "wet method" of cement production. This process involved the creation of a water-based slurry of crushed cement ingredients that is then introduced, after pre-heating and pre-calcining, into a bituminous coal / hazardous waste-fired rotary cement kiln to produce the cement "clinker" base used as the primary uncrushed component of the final cement product. The wet process, used traditionally in cement manufacturing since its development, was more energy-intensive as additional energy was required to drive off the water contained in the slurry. Given developments in dry crushing technology to ensure more uniform and consistent crushed mixes, however, the wet method has been generally replaced with a "dry method" that eliminates the need for water while maintaining the quality of the cement product. The Keystone Cement facility converted its facility process from two wet method rotary kilns to a single, larger and more energy-efficient dry method rotary kiln and associated pre-heater and pre-calciner in 2009.

Since the conversion, the Keystone Cement facility initially had difficulty maintaining compliance with an established source limit for NO_x. Required Continuous Emissions Monitors ("CEMs") for NO₂, SO₂ and Opacity monitor these emissions for compliance purposes. Corrective action by the company following a compliance settlement for previous violations is intended to ensure continued compliance with the permitted emissions limits from the subject sources.

B. Ambient Air Particulate Monitoring Site near Keystone Cement

In response to regional concerns about pollution arising from the Keystone Cement facility and impacting the local community of Bath, in December of 1995, High-Volume Total Suspended Particulate ("Hi-Vol TSP") ambient air samplers were located on the campus of the George Wolf Elementary School located on 300 Allen Street in Bath Borough. The samplers were configured to collect one 24-hour sample every day over a four-day period before requiring servicing. In November 2004, a fifth Hi-Vol TSP sampler was added. This additional sampler helped to ensure improved sample collection efficiency by allowing an additional day for field staff to retrieve collected samples and to reset the monitors to collect for the next five days. All five Phase I monitors were operated until late 2013 or early 2014. Table 1 summarizes the monitoring location, the Phase I sampling monitors, and their sampling schedule.

⁴ EPA Technology Transfer Network – AP-42 Chapter 11.6 – Portland Cement Manufacturing
<https://www3.epa.gov/ttnchie1/ap42/ch11/final/c11s06.pdf>

Table 1 - Phase I Monitoring Near Keystone Cement Co.

Table 1 - Phase I Monitoring Near Keystone Cement Company				
Sampling Location				
Monitoring Point:	32K			
Address:	300 Allen St, Bath PA 18014			
County	Northampton			
Municipality	Borough of Bath			
DD Lat	40.72344		N	
DD Long	-75.38932		W	
Sampler Information				
Type:	Hi-Vol TSP			
Filter:	Glass			
Analysis Code (SAC):	3IV			
Sampling Duration:	24-hr			
Sampler ID	Start Date	End Date	Schedule*	Total Sampling Period (Yr)
32KB	12/5/1995	1/15/2014	1:4 / 1:5	18.1
32KC	12/6/1995	1/16/2014	1:4 / 1:5	18.1
32KD	12/7/1995	1/17/2014	1:4 / 1:5	18.1
32KE	12/4/1995	12/29/2013	1:4 / 1:5	18.1
32KF	11/19/2004	1/14/2014	1:5	9.2
* 4 Monitors operated on a 1 sample in 4 day schedule (1:4) until Nov 2004. All samplers operated on a 1:5 schedule in 2005 until 2014				

Figure 1 is an aerial overview of the sampling area showing the samplers in relation to the Keystone Cement facility and the Borough of Bath.

Figure 1 - Sampling Location for Keystone Cement Monitoring 1995 - 2015



Over the 18-year period of Phase I, the monitoring configuration provided daily (24-hour average) ambient levels of TSP, sulfate and iron that could be present in the community. Additionally, the analysis included toxic metals; however, the investigative analysis method used for the samples had toxic metals reporting limits that, while sufficient to screen for potential unhealthful acute and higher-level chronic concentrations, were insufficient to evaluate lower concentrations that could possibly contribute to smaller incremental chronic lifetime cancer risk. After consultation with Central Office Bureau of Air Quality staff in 2012, the monitoring was modified by the NERO staff to:

1. reduce the number of monitors, and therefore the sampling frequency, to collect data on a statistically representative schedule (i.e., 1 sample every 6 days) instead of daily, and
2. collocate a monitor using more precise toxic metals investigative methods to better ascertain, at a screening level, if monitored toxic metals pose a chronic inhalation health risk or hazard at the monitored location.

The Phase II modified monitoring plan commenced on January 23, 2014, and completed on March 31, 2017. Table 2 summarizes the Phase II monitoring configuration and schedule. Note that in 2015, third-party data quality assurance capability was not available. All data following the “End Date (QA)” date for the Phase II sampling is provisionally quality assured pending Quality Assurance checking from the Quality Assurance and Analysis Section of the Air Quality Monitoring Division.

Table 2 - Phase II Monitoring Near Keystone Cement Co.

Table 2 - Phase II Monitoring Near Keystone Cement Company						
Sampling Location						
Monitoring Point:		32K				
Address:		300 Allen St, Bath PA 18014				
County:		Northampton				
Municipality:		Borough of Bath				
DD Lat:		40.72344 N				
DD Long:		-75.38932 W				
Sampler Information						
Type:		Hi-Vol TSP				
Filter:		Glass or Quartz				
Analysis Code (SAC):		3IV or 3IC				
Sampling Duration:		24-hr				
Sampler ID	SAC Code - Filter Type	Start Date	End Date (QA)	End Date (Prov)	Schedule*	Total Sampling Period (Yr)
32KB	3IV - Glass	1/23/2014	5/24/2015	11/26/2015	1:6	1.8
32KC	3IC - Quartz	1/23/2014	7/17/2015	3/31/2016	1:6	2.2
* Conforms to U.S. EPA NAAQS Ambient Monitoring Network schedule						

C. Comparison of Analytical Methods

Table 3 summarizes the two DEP lab analytical methods used (3IV and 3IC) and the average lab reporting limits for the results. The toxics 3IC analysis used in Phase II sampling has significantly lower reporting limits, thus enabling more accurate and precise measurement of lower concentrations of toxic metals over time. Measurement at lower concentrations allows for better assessment of concentrations that could be considered unhealthy chronic exposure levels (i.e., more than one year).

Details on the particulate collection and filter analysis methods can be found on EPA's Technology Transfer Network at <https://www3.epa.gov/ttn/amtic/inorg.html>. Phase II TSP collection methods were performed in accordance with EPA Toxic Inorganic Compendium method IO 2.1 (*Sampling of Ambient Air for Total Suspended Particulate Matter (SPM) and PM₁₀ Using High Volume (HV) Sampler*) with samples analyzed in accordance with EPA Toxic Inorganic Compendium method IO 3.5 (*Determination of Metals in Ambient Particulate Matter Using Inductively Coupled Plasma/Mass Spectrometry (ICP/MS)*).

Table 3 - Summary and Comparison of Monitoring Lab Analysis Suites

Table 3 - Summary and Comparison of Monitoring Lab Analysis Suites				
3IV - Hi-Vol Investigative Ambient Air Analysis				
Analyte	CAS No.	Lab Test Code	Reporting Limits (RL)	
			Mass on Filter (ug)	Mass/Vol (ug/m3)
Arsenic	7440-38-2	01002E	14.4	0.0073
Sulfate Ion	18785-72-3	00945E	N/A	N/A
Cadmium	7440-43-9	01027E	36	0.019
Chromium, Total	7440-47-3	01034E	18	0.0094
Lead (non-regulatory)	7439-92-1	01051E	72	0.038
Iron	7439-89-6	01045E	180	0.093
Nickel	7440-02-0	01067E	72	0.039
Zinc	7440-66-6	01092E	144	0.072
Particulate by Weight on Air Filters	N/A	WEIGHT	N/A	N/A
RL for Mass/Volume calculation is the average of reported sample RL's from 1994 through 2013 at Bath monitoring site.				
3IC - Hi-Vol Investigative Concentrated Ambient Air Analysis				
Analyte	CAS No.	Lab Test Code	Reporting Limits (RL)	
			Mass on Filter (ug)	Mass/Vol (ug/m3)
Arsenic	7440-38-2	01002F	1.44	0.0007
Beryllium	7440-41-7	01012F	0.36	0.00023
Cadmium	7440-43-9	01027F	0.036	0.00023
Chromium, Total	7440-47-3	01034F	7.2	0.0046
Lead (non-regulatory)	7439-92-1	01051F	7.2	0.0047
Manganese	7439-96-5	01055F	0.72	N/A*
Nickel	7440-02-0	01067F	3.6	0.0046
Zinc	7440-66-6	01092F	7.2	N/A*
Particulate by Weight on Air Filters	N/A	WEIGHT	N/A	N/A
RL for Mass/Volume calculation is the average of reported sample RL's from 1/23/14 through the duration of monitoring in 2015.				
* Mn and Zn were detected on all 3IC samples therefore no average reporting limit is estimated.				

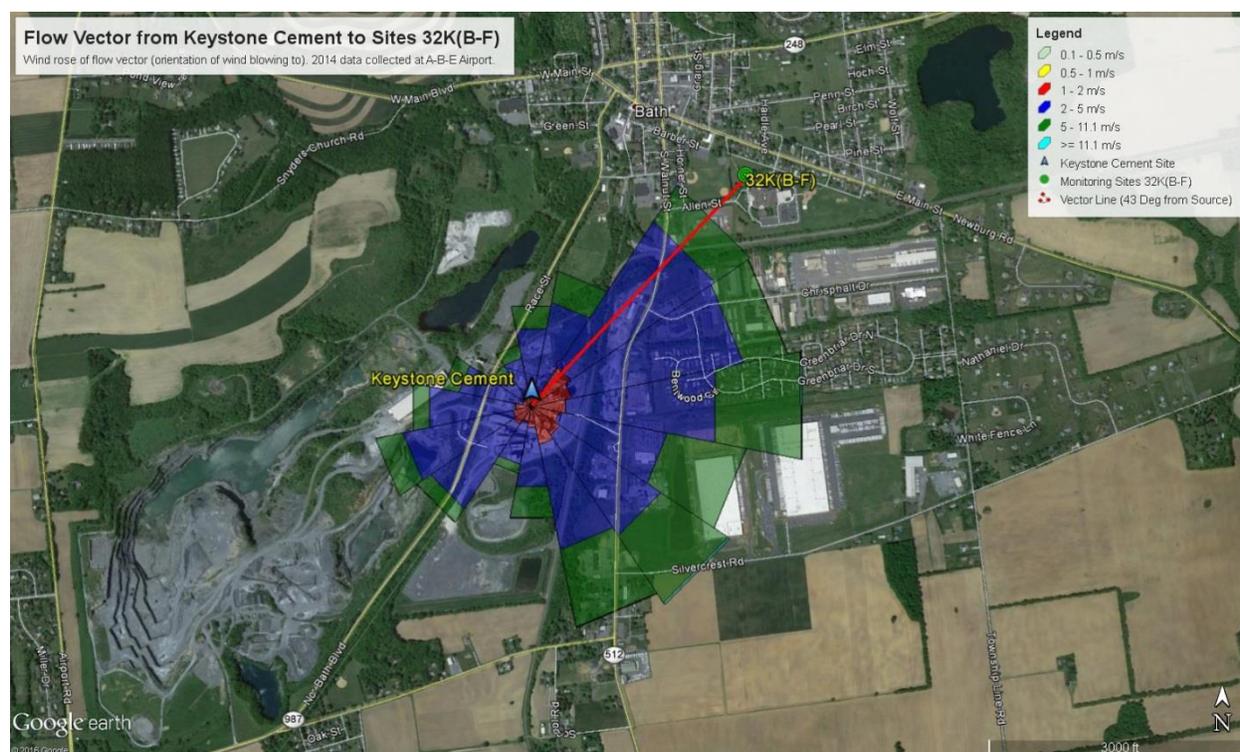
III. Phase I Sampling at Site 32K

A. Phase I Data Collection

From December 4, 1995 through January 17, 2014, 5,541 valid 24-hour TSP samples were collected out of a potential 6,619 samples, yielding a site data availability rate of 84%. With daily sampling, this data availability rate is sufficient to estimate annual mean concentrations of TSP. Phase I filters were weighed for TSP and analyzed for sulfates and a suite of metals using lab analysis code (SAC) 3IV.

The samplers continue to be located in an area of downwind influence from the source as evidenced by an examination of directional wind data from a nearby location with similar meteorological and terrain features. Wind speed and directional data from the commercial Lehigh Valley International Airport (“ABE”) located approximately 5 miles to the southwest of the facility was used as a surrogate for on-site meteorological data collection. Figure 2 shows the ABE airport wind data as a wind rose reflecting the flow vector of the wind. The flow vector points in the *direction towards which a recorded wind is blowing* (the “blowing to” direction). The data indicate that the monitors were located in a representative area downwind of the facility as the monitors were located within a portion of the Keystone Cement facility’s predominate downwind vector.

Figure 2 - Keystone Cement Wind Flow Vector Map



Because the sampling site is located in a largely downwind vector from the source and is relatively close to the source, it would be expected that the samplers would be capable of capturing the effects of particulate emissions from the Keystone Cement facility and measuring their relative changes over time. This would be evidenced in the data by discernable step changes in ambient particulate concentration trends coinciding with known particulate emission changes at the facility.

B. Phase I Data Analysis

Phase I data consists largely of measurements of TSP, sulfate and iron. These analytes were mostly detected in each sample in varying concentrations. Table 4 summarizes the annual and three-year averages (arithmetic mean) and the annual and three-year average percent changes for these pollutants for each year from 1996 through 2013. Additionally, percent data availability for each quarter of each year for all samplers is included. Annual, three-year and three-year trend average percent change decreases greater than 5% are denoted in green, while increases greater than 5% are indicated in red.

In general, annual data availability was suitable for TSP and sulfate annual average calculation because annual data availability percentages were close to or greater than the recommended minimum of 75% valid samples per quarter, for each quarter, of the possible 365 or 366 samples collected over a given year⁵. On an annualized basis, TSP and sulfate had greater than 75% data availability for all years.

Annual data availability for iron was slightly less than the 75% minimum for 2012. This was due to a higher rate of invalid iron sample results due to apparent analysis errors. However, given the daily nature of the sampling and a data availability of 71%, the 2012 annual average for iron can be reasonably estimated for comparison purposes.

Figure 3 plots the measured TSP concentrations in $\mu\text{g}/\text{m}^3$ from all five monitors (32KB-32KF) from December 4, 1994 to January 7, 2014. Figure 4 plots one monitor (32KB) for the same time period. In both figures, it appears that sometime in the fall of 2008, the overall observed concentrations of TSP began a downward trend. This observation is easier to discern in Figure 4.

⁵ Of the 72 possible quarters of data, only eight (11%) of the quarters were below the 75% minimum quarterly data availability requirement. Of those eight, five were at 72% - 73% and two were in the mid to high 60th percentile. These quarters were retained for annual average calculation. The lowest data availability percentage quarter was 3rd quarter 2008 at 48%. This quarter was conservatively retained for 2006 annual average calculation, as that quarter's average TSP concentration was the highest ($48.5 \mu\text{g}/\text{m}^3$) quarterly average observed in the 18 years of monitoring across all quarters. The years 1996, 2000, 2004, 2008 and 2012 were Leap Years with an extra day in the 1st quarter and 366 possible samples for the year.

Figure 3 – Monitoring Point 32K - TSP Concentrations - All Samplers - 1995-2013

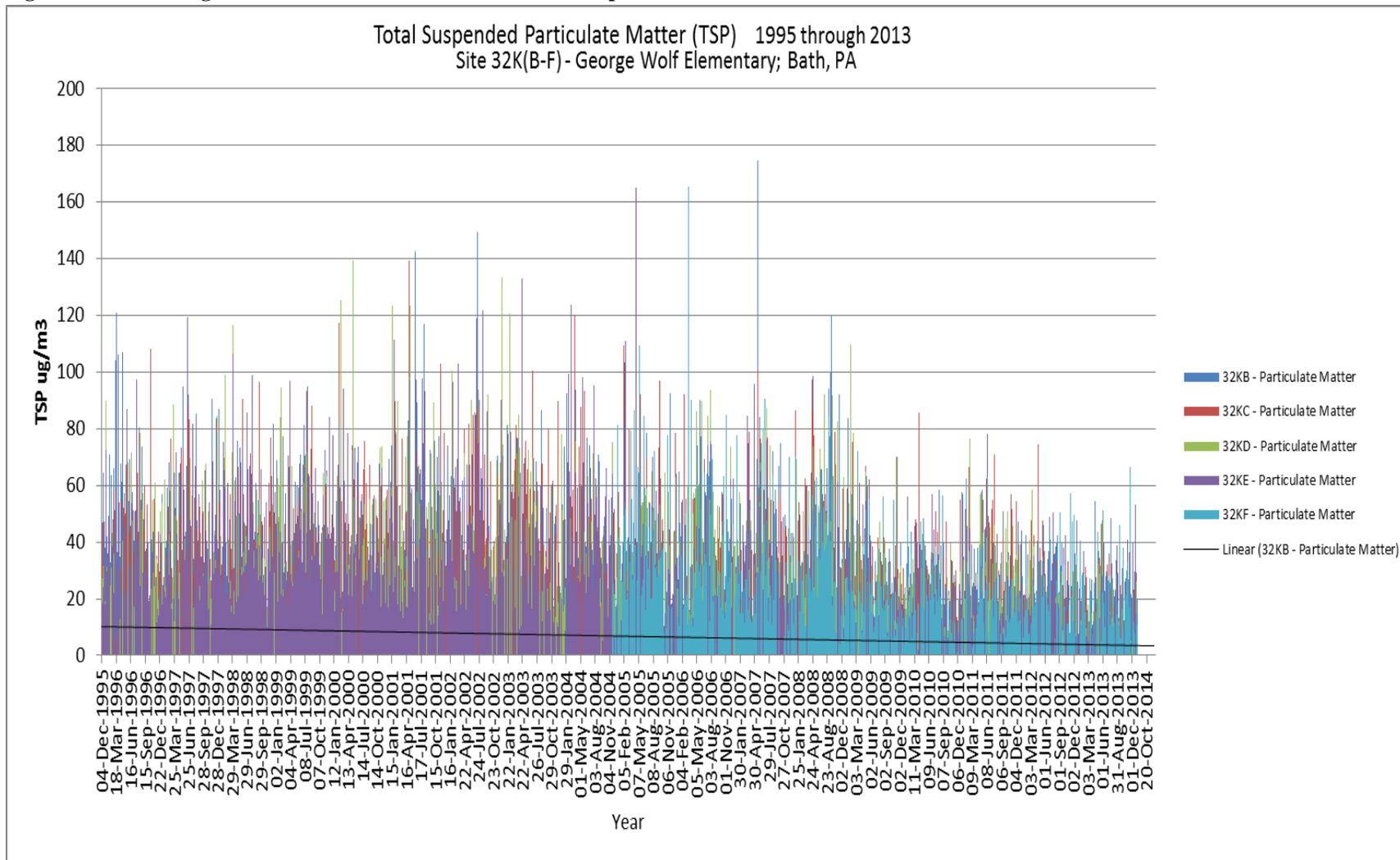
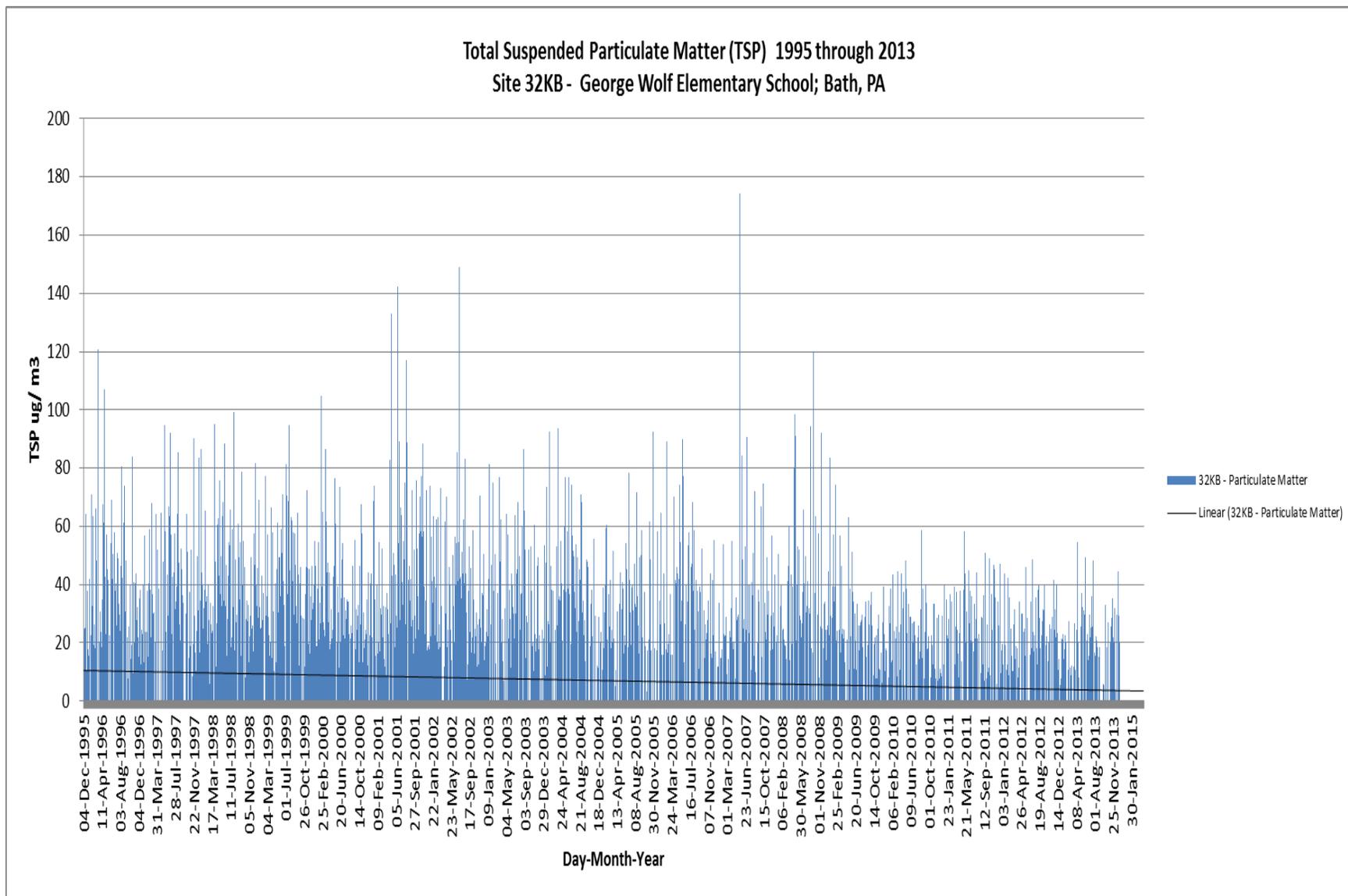


Figure 4 – Monitoring Point 32K - TSP Concentrations - Sampler 32KB - 1995-2013



Annual TSP, sulfate and iron average concentrations in $\mu\text{g}/\text{m}^3$ are plotted in Figures 5 and 6. An apparent downward trend in sulfate concentrations begins in 2008/2009 and generally corresponds to the TSP reductions.

Figure 5 - Annual TSP and Sulfate Trends at Monitoring Point 32K

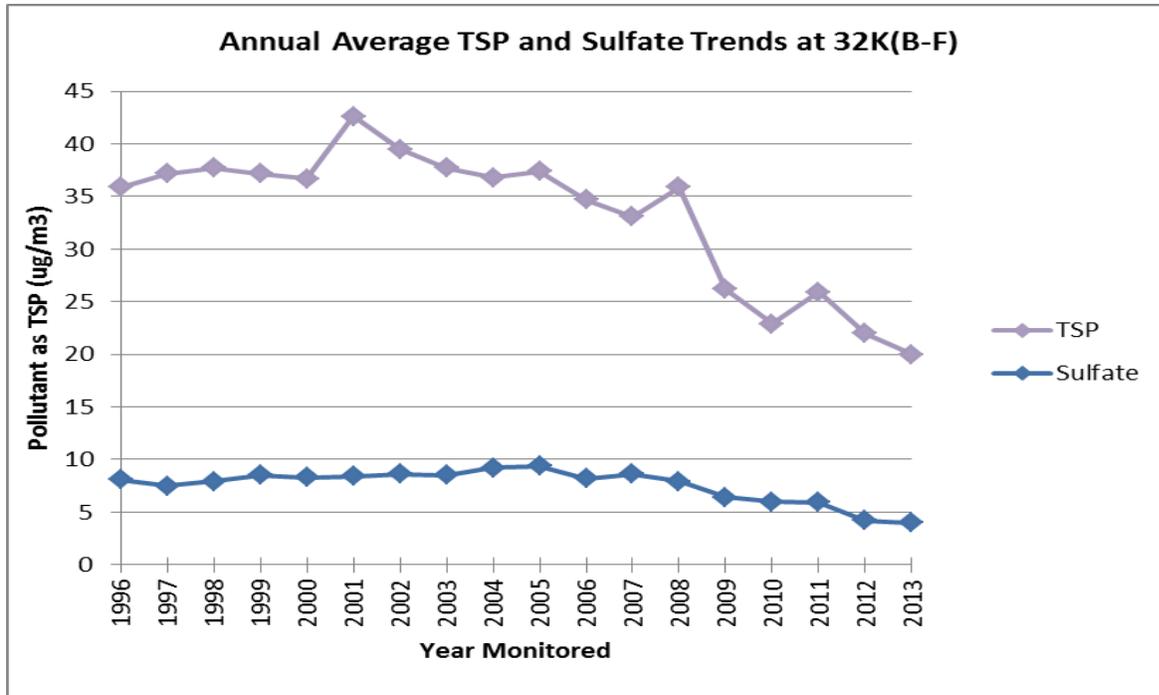
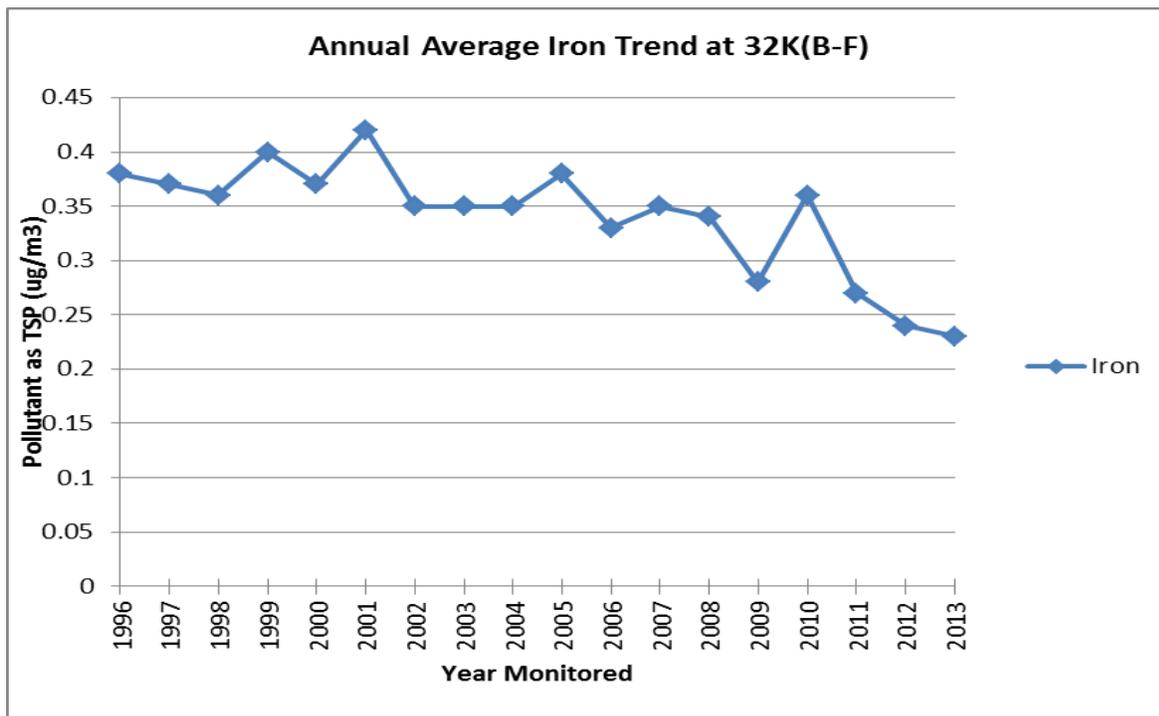


Figure 6 - Annual Average Iron Trend at Monitoring Point 32K



Note that the average ambient air iron concentrations for 2010 are at levels corresponding to, or higher than, years when the Keystone Cement facility was operating using the wet method. The iron data for this year had a higher frequency of values in the upper range of the historical distribution of data. This could be related to specific activities at the facility or possibly another localized intermittent, temporary source. However, after the 2010 “spike,” observed concentrations of iron at the monitoring point continue the generally downward linear trend from 2008 through 2013.

In order to gauge a relative “significant” increase or decrease in monitored data, a simple screening method of comparison is to compare the three-year averages of two three-year periods. If an increase or decrease is greater than or equal to 15%, the change between the two three-year periods is considered significant. The three-year averaging period for the two periods is done to smooth potential variation in monitored concentrations that can occur within a period of less than three years (e.g., temporary plant shutdowns or modifications, localized construction, sampling equipment issues, etc.). Changes less than 15% over time may also be “significant” depending on their duration as it may reflect larger regional trends.

Table 5 summarizes the mean three-year average concentration data observed for the three-year period 2006 through 2008 and the three-year period 2009 through 2011. This division of the three-year averaging periods best reflects the times representing the physical changes made at the Keystone Cement facility to convert from the “wet” to “dry” Portland cement manufacturing process.

Table 5 - Wet Method (2006 – 2008) and Dry Method (2009-2011) 3-yr. Average (Mean) TSP / Sulfate and Iron Concentration Percent Differences

3 Yr Average Pollutant Concentration Trend for Monitoring Point 32K (from Table 4)			
Pollutant:	TSP (ug/m3)	Sulfate (ug/m3)	Iron (ug/m3)
3 Yr Average 2006-2008	34.57	8.23	0.34
3 Yr Average 2009-2011	25.00	6.10	0.30
% Difference	-28%	-26%	-11%

The monitoring data percent reductions summarized in Tables 4 and 5 in conjunction with the daily particulate data displayed in Figures 3 through 6 show that, beginning in 2008 and 2009, observed ambient concentrations of TSP and sulfate decreased significantly⁶. Reduction in the

⁶ For this report, a “significant decrease” is a decrease between two contiguous three-year concentration averages greater than 15%. The first six-year trend reduction 15% or greater was in 2010 for TSP (19% reduction), 2009 for sulfate (15% reduction) and 2013 for iron (24% reduction).

average ambient concentration of iron over the six-year period was less significant but consistent with an overall reduction trend beginning in the 2004/2005 time period.

Comparison to Health-Based Standards for Ambient Air

Total Suspended Particulate (TSP)

There is no national ambient air monitoring standard for TSP. The National Ambient Air Quality Standards (NAAQS) for particulate matter are for the PM₁₀ and PM_{2.5} (particulate matter 2.5 microns or less) particulate fractions of the TSP in ambient air. Both the highest annual TSP concentration from 1996 through 2013 (42.6 µg/m³ in 2001) and the highest three-year average (39.9 µg/m³ for 2001 – 2003) are well below the 150 µg/m³ three-year annual average PM₁₀ NAAQS standard. In addition, the TSP values are below a more protective 50 µg/m³ 24-hour average health-based PM₁₀ standard set by the CARB for California.

Insufficient data exist from the TSP data alone to accurately estimate the PM_{2.5} fraction of the TSP. A conservative estimate of 50% PM_{2.5} fraction TSP for 2011-2013 would yield a three-year averaged PM_{2.5} estimate of 11.9 µg/m³. This is less than the current 2012 PM_{2.5} NAAQS of 12.0 µg/m³ averaged over three years.^{7,8}

Sulfate (SO₄²⁻)

Besides being directly emitted from combustion sources and entrained in the air from soils and dust created from sulfate-bearing materials, sulfate is also an important secondary aerosol arising from atmospheric photochemical interactions with sulfur dioxide (SO₂). In addition to its potential harm to human health, it contributes to reducing visibility and increasing acidification through wet deposition. It is a participant in the formation of cloud-condensing nuclei and can influence cloud physics. Because of its ability to absorb upper-atmospheric shortwave radiation, it is also a contributor to climatic cooling. (Hand, et al. 2012)⁹

Ambient concentrations of sulfate in the air fluctuate seasonally as summertime weather typified by higher solar insolation, more frequent stagnation events, and higher relative humidity lead to higher atmospheric peak concentrations than in fall/winter months. This oscillation is evident in plotted data as shown in Figure 7. Concentrations of sulfate greater than 10 µg/m³ are mostly during the spring/summer while concentrations less than 10 µg/m³ are observed more frequently in the fall/winter.

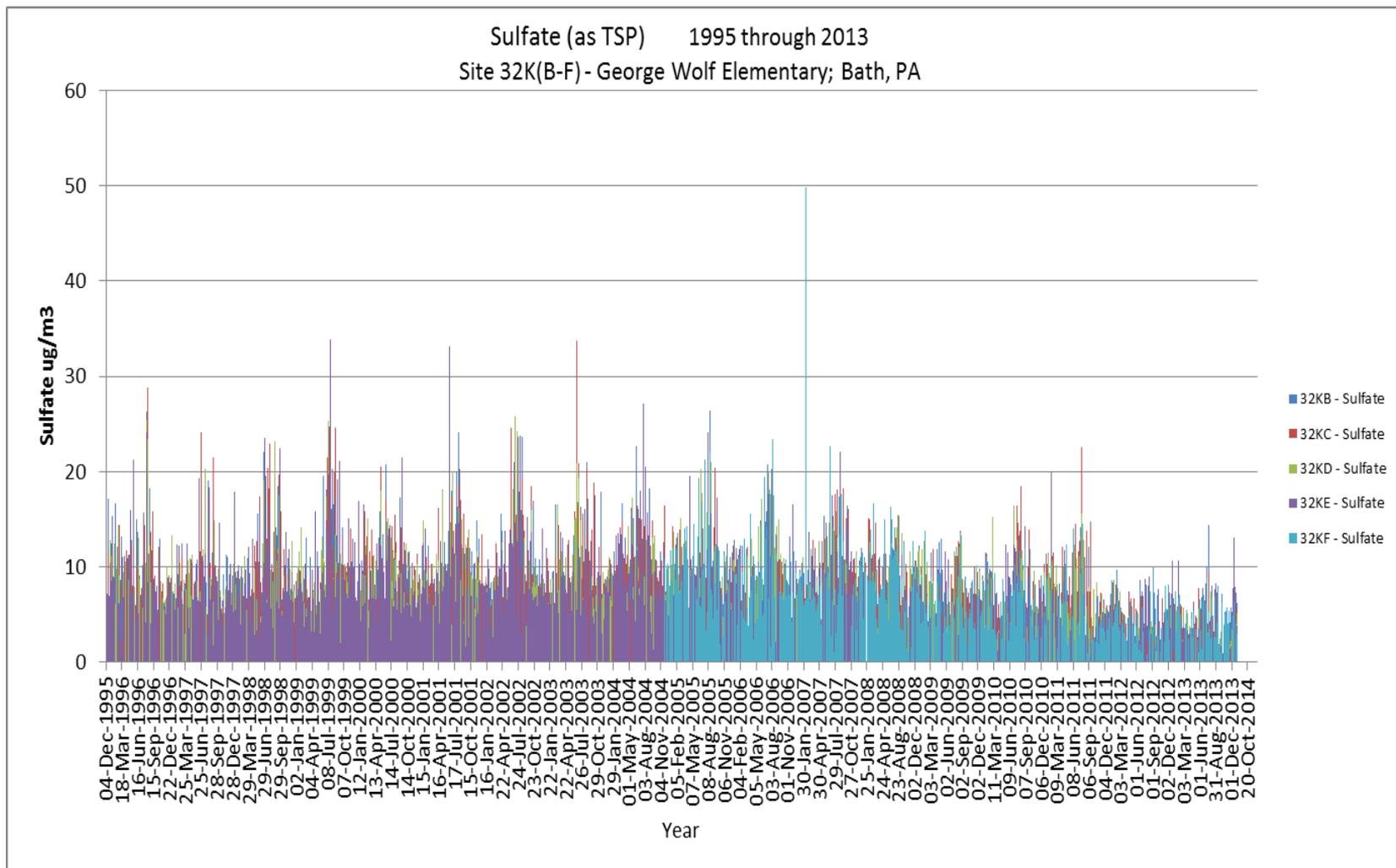
The health impacts from sulfate are largely reflected in its being a significant component of PM_{2.5} mass (30-60% in the eastern United States). Regulation of PM_{2.5} and PM₁₀ in areas impacted by sulfate is the primary tool used to reduce ambient air sulfate concentrations.

⁷ The Borough of Bath, Northampton County, as part of the Allentown-Bethlehem-Easton Air Basin, is in demonstrated attainment of the 2012 PM_{2.5} NAAQS. It is important to note that estimation of PM_{2.5} concentrations from TSP sampling is not an approved reference method for estimating PM_{2.5} in ambient air and is included for informational purposes only.

⁸ The annual CARB PM_{2.5} standard is the same as the federal PM_{2.5} NAAQS.

⁹ [J.L. Hand et al.: Particulate sulfate ion concentration and SO₂ emission trends in the United States from the early 1990s through 2010. Journal of Atmospheric Chemistry and Physics, Vol 12, 10353-20365, 2012](#)

Figure 7 - 24-Hr Sulfate Concentrations at Site 32K (B-F) from 1995 through 2013



There is no NAAQS for sulfate. However, CARB has a 24-hour average standard for sulfate in PM₁₀ at 25 µg/m³. Historically, ambient 24-hour concentrations of sulfate in TSP at the site exceeded 25 µg/m³ on only 13 occasions over the 5,617 days (18.1 years) sampled, or 0.23%. The dates and 24-hour concentrations are summarized in Table 6.

Table 6 – Observations of Sulfate Concentration Greater than 25 µg/m³ at Monitoring Point 32K

Sample Date	Ranked TSP Sulfate Concentration (ug/m3)
2/8/2007	49.8
7/18/1999	33.8
6/26/2003	33.8
6/14/2001	33.1
7/19/1999	30.3
8/7/1996	28.8
7/22/2004	27.1
7/17/1999	27.1
8/13/2005	26.4
8/6/1996	26.3
7/2/2002	25.9
8/4/1996	25.5
7/4/1999	25.4

Except for the highest reading on February 8, 2007, all other values over 25.0 µg/m³ sulfate were observed during a period from mid-May to mid-August and did not exceed 34 µg/m³. Assuming that 60% of the sulfate in the TSP sample is PM₁₀, the resulting concentration (20.4 µg/m³ max.) is less than the 25 µg/m³ 24-hour ambient standard set by CARB. The same period of occurrence for the values corresponds with the seasonal oscillation of sulfate in ambient air.

The highest value of 49.8 µg/m³ is 44% higher than the 2nd highest value and is observed in February, a period typically characterized by low ambient sulfate concentrations. Additionally, TSP and other metal concentrations for that day were not disproportionately higher than the day before and after the high-value day. This lends to two possibilities: 1) the value is a potential analytical error as the real presence of comparatively high concentrations of sulfate in the air without some corresponding increase in mass collected on the filter is unlikely, or 2) the sampler collected material during the 24 hours that was composed of considerably higher sulfate concentration per unit total mass than is typical.

Iron (Fe)

Particulate matter containing iron can be found in ambient air. Iron is generally measured as a fraction of the crustal component of PM_{2.5} that can be introduced from entrainment from the ground surface or from the crushing and handling of stone containing concentrations of iron (e.g.

pyritic shales and some limestones). Combustion sources using coal, iron and steel foundries, and ferro-metallic processing and handling, are the most common source categories for primary iron and iron-bearing particulate emissions.

Iron is also a critical component of Portland cement manufacturing. Along with oxides of aluminum and magnesium, iron serves as a flux in the Portland cement kiln process to lower the formation temperature of the calcium silicates that comprise the majority of the cement clinker. Iron can also help impart some strength to the final product. Iron can be introduced to the kiln from the addition of scrap iron, iron-bearing (pyritic) shales or fly ash from coal combustion, among other methods. It also may be naturally present in the limestone used to make the cement.

Figure 8 shows the daily 24-hour ambient air iron concentrations observed from the five samplers at site 32K from late 1995 through 2013. With the exception of two samples, all concentrations were below $2.0 \mu\text{g}/\text{m}^3$.

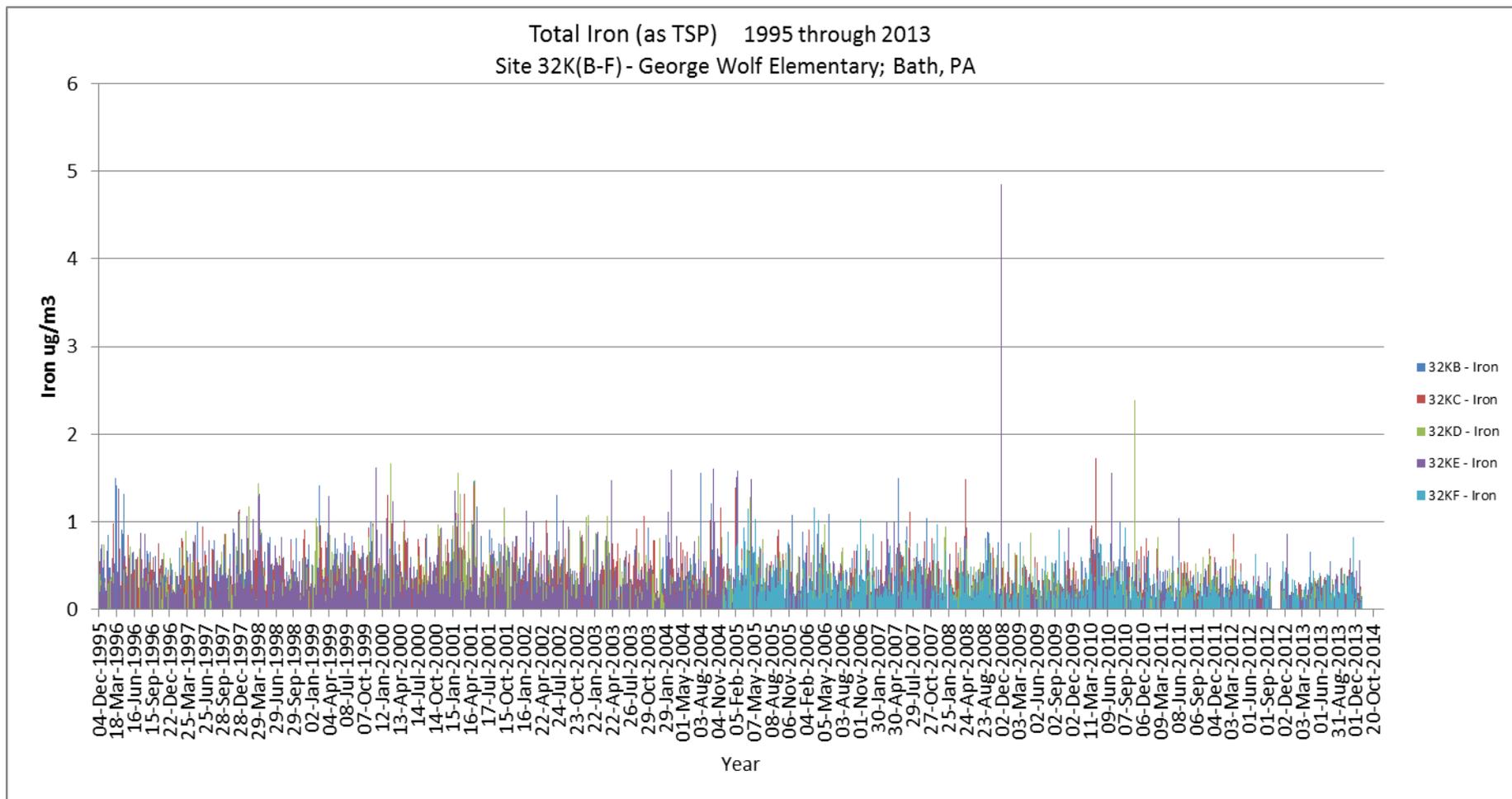
Although ingested iron in very high concentrations above its nutritive value can lead to toxic side effects, there is no reported toxicity specifically associated with the inhalation of iron. It is not considered a hazardous air pollutant. As a particulate, iron's presence in ambient air is reduced as overall particulate matter is reduced. It is monitored in speciated $\text{PM}_{2.5}$ samples.

There is no health-based ambient air standard for iron.

C. Conclusions from Phase I Monitoring: 1995 through 2013

- The TSP samplers located at Site 32K were sufficiently located and operated to collect near-continuous daily ambient air TSP, sulfate and iron concentrations from 1995 through 2013.
- Available meteorological data (wind speed and direction) used to site monitors at the Keystone Cement facility demonstrated that Site 32K was in a portion of a predominant downwind vector from the facility.
- TSP concentrations at the site dropped by 28% averaged over the three years following a major conversion at the Keystone Cement facility from a wet to dry Portland cement method in 2009. Concentrations of sulfate and iron in the ambient air dropped by 26% and 11%, respectively, over the same periods.
- No exceedances of applicable NAAQS for particulate matter were observed. Thirteen values of sulfate were observed over a CARB sulfate ambient standard of $25 \mu\text{g}/\text{m}^3$. However, given that the CARB standard is for the PM_{10} fraction (not TSP as measured at 32K), it is unlikely that health was adversely affected because a significant portion of TSP is non-respirable.
- Due to limitations in reporting limits of the filter analytical method (SAC 3IV), insufficient data exists to appropriately conduct a health-based risk/hazard screening for toxic metals. Phase II monitoring focuses on toxic metals using an analytical method with lower reporting limits to help assure better estimates of mean annual concentrations suitable for screening potential chronic exposure effects.

Figure 8 - 24-Hr Iron Concentrations at Site 32K (B-F) from 1995 through 2013



IV. Phase II Sampling at Site 32K

A. Phase II Data Collection

Beginning on January 23, 2014, three of the five existing Phase I TSP samplers were deactivated. The two remaining samplers, 32KB and 32KC, were placed on a systematic 1-in-6-day schedule. The original Phase I analysis code 3IV was continued on glass TSP filters to provide data continuity from the Phase I monitoring. Quartz TSP filters were run on sampler 32KC to sample for concentrated metals analysis using analysis code 3IC which is more suited to trace metals analysis. The sampling frequency for both 32KB and 32KC aligned with the national EPA schedule for 1-in-6-day sampling for criteria pollutant compliance monitoring.

A summary of the sampling activity for Phase II is presented in Table 7.

Table 7- Summary of Phase II Sampling at Monitoring Point 32K

Table 7 - Phase II Monitoring Near Keystone Cement Company						
Sampling Location						
Monitoring Point:	32K					
Address:	300 Allen St, Bath PA 18014					
County:	Northampton					
Municipality:	Borough of Bath					
DD Lat:	40.72344 N					
DD Long:	-75.38932 W					
Sampler Information						
Type:	Hi-Vol TSP					
Filter:	Glass or Quartz					
Analysis Code (SAC):	3IV or 3IC					
Sampling Duration:	24-hr					
Sampler ID	SAC Code - Filter Type	Start Date	End Date (QA)	End Date (Prov)	Schedule*	Total Sampling Period (Yr)
32KB	3IV - Glass	1/23/2014	5/24/2015	11/26/2015	1:6	1.8
32KC	3IC - Quartz	1/23/2014	7/17/2015	3/31/2016	1:6	2.2
* Conforms to U.S. EPA NAAQS Ambient Monitoring Network schedule						

In mid-2015, independent Quality Assurance for air toxics samples did not occur due to the lack of staff in the Quality Assurance and Analysis Section. Data collected from the “End Date (QA)” to the end of sampling at the location was provisionally quality assured by the Toxics Monitoring Section prior to this analysis. Therefore, the quality of the provisional data has not been independently verified by Quality Assurance staff.

During the Phase II sampling, availability of data was limited due to issues with the samplers collecting outside of the acceptable bounds of flow changes over a given 24-hour sampling period (allowable flow change of +/- 2.0 CFM over 24 hours). This reduced the quarterly data availability to below the recommended 85% available data per quarter that, in turn, reduces the certainty of estimates of mean concentrations that are needed to perform health-based chronic health screening.

The data availability for the Phase II sampling is summarized in Table 8. Quarterly percent Data Availability (%DA) above the 85% recommended percentage is highlighted in green.

Table 8 - Phase II Data Availability Summary

Phase II Monitoring Quarterly and Annual Data Availability Percentage by Sampler and Year											
Sampler by Year		Quarter								Annual	
		1Q		2Q		3Q		4Q			
2014	Potential Samples	15		15		15		16		61	
	Sampler	# Valid	% DA								
	32KB	13	87%	12	80%	8	53%	10	63%	43	70%
	32KC	14	93%	9	60%	7	47%	11	69%	41	67%
2015	Potential Samples	15		15		15		15		60	
	Sampler	# Valid	% DA								
	32KB	7	47%	11	73%	13	87%	10	67%	41	68%
	32KC	9	60%	10	67%	15	100%	15	100%	49	82%
2016	Potential Samples	16									
	Sampler	# Valid	% DA								
	32KC	13	81%								

Given the incomplete quality of the data for either given year, the use of average (arithmetic mean) values to estimate a mean pollutant concentration for health-based toxic metals screening may underestimate potential chronic risks and hazards associated with lifetime exposure. The use of more conservative, alternative methods for choosing a health-based screening concentration is recommended.

B. Phase II Data Analysis

1. Collocation of Phase II Samplers

The sampler 32KB was operated to collect representative data for comparison to the Phase I results. If TSP sampling was consistent between both phases, the Phase II 32KB sampler results should be similar in magnitude and trend to late Phase I 32KB results. If the data appear to indicate such similarity, the Phase II data from 32KB can be used to monitor for any unusual events occurring during the concurrent toxics metals sampling on sampler 32KC.

As shown in Figure 9, the pattern of TSP concentrations collected from 32KB do not appear to shift in such a way to suggest that TSP concentrations between phases had changed due to changes in the sampling design. Additionally, the sulfate and iron concentrations from both phases are similar¹⁰.

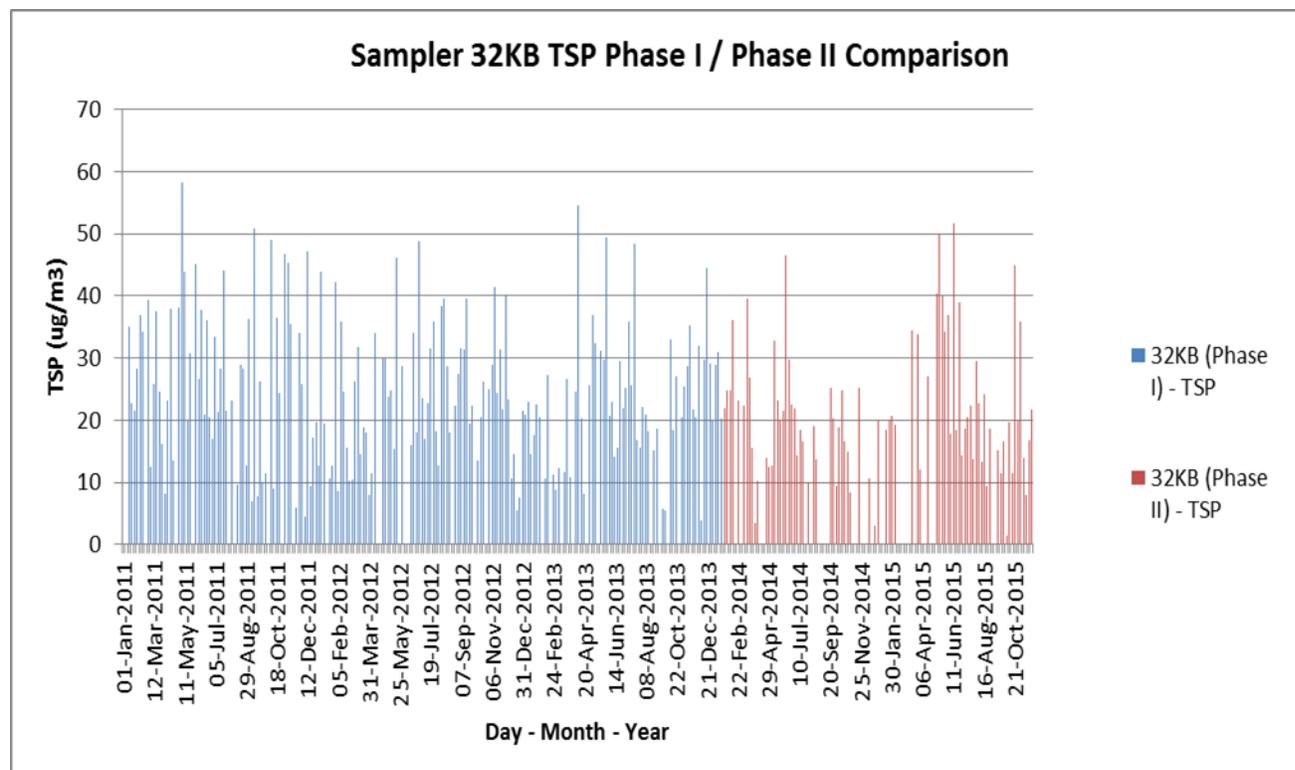
During Phase II sampling, the 32KB sampler did not sample any 24-hour concentration of TSP, sulfate or iron that would be indicative of an exceptional event. All sampled Phase II concentrations of TSP, sulfate and iron were within the observed distribution of all the samples collected over the 18 years of Phase I sampling.

A comparison between arithmetic mean (average) TSP values for 32KB and 32KC operating simultaneously is not appropriate as data availability was insufficient over multiple quarters to make such comparisons with confidence without resulting to more advanced statistical analyses. However, the average TSP concentrations for a period of simultaneous 1-in-6-day sampling from

¹⁰ Graphical data for sulfate and iron are included in the attachments to this document. Observed similarities of TSP, sulfate and iron have not been validated through statistical analysis of the data. That analysis is beyond the scope of the report; however, the similarities in the graphs at the point of transition are rather evident.

January 5, 2014 through November 26, 2015, varied by less than 15% ($21.8 \mu\text{g}/\text{m}^3$ vs. $24.8 \mu\text{g}/\text{m}^3$) between the samplers.

Figure 9 - Sampling Phase Comparison of TSP Data



2. Toxic Metals Risk / Hazard Screening

The primary purpose of the Phase II sampling was to collect sufficient valid data to compare an estimate of the mean concentration of toxic metals over the period monitored to health-based pollutant concentration screening values. This is done in the attempt to determine if additional investigation is needed out of a concern for a *potential* unacceptable increase in exposed population lifetime cancer incidence or chronic inhalation hazard. Toxic metals that are observed to have estimated mean concentrations greater than the health-based screening value should be further investigated and not necessarily assumed to be unhealthy until the additional investigation is complete.

Typical sampling for toxic metals (SAC 3IC) includes a variety of metal analytes varying from little to no toxicity (zinc¹¹) to high relative toxicity (lead and arsenic). Refer to Table 3 for additional details on analytes and reporting limits for the 3IC test.

EPA guidance for the screening of ambient air metals concentration data for potential health-based effects recommends that sampled metals only be screened if the non-detect percent frequency

¹¹ Zinc, of all the metals in SAC 3IC, is not a hazardous air pollutant. It has been traditionally included in monitoring suites for decades as it is associated with a variety of metals extraction, processing and manufacturing industries and is detected at a higher rate of frequency than most other toxic metals, thus providing a potential data surrogate for source investigative purposes.

(%ND) is less than 85%. In other words, more than 15% of the total collected valid samples should have a reported detection above the reporting limit for that metal to be included in the screening.

Table 9 summarizes the percentage rate of non-detected data for the 92 valid samples (75% of total) collected from January 29, 2014 through January 31, 2016. Metals suitable for screening purposes are highlighted in green.

Table 9 – 32KC Phase II Percent Non-Detect (% ND) by Analyte

Phase II Sampling Metals Data Availability and % Non-Detection (% ND) Rates			
Analyte	# Valid Samples	# ND	%ND
Manganese	92	0	0%
Zinc	92	0	0%
Arsenic	92	32	35%
Lead	92	52	57%
Cadmium	92	61	66%
Chromium (Total)	92	88	96%
Nickel	92	91	99%
Beryllium	92	92	100%
Total Potential Samples:		123	
Total % Data Availability:		75%	

3. Determining Screening Concentrations

To screen ambient air toxics data against health-based values, an estimate of a mean concentration of the measured pollutants to which a population might be exposed over a lifetime must be calculated to serve as a *screening concentration*. The screening concentrations are then compared to health-based screening values. If the screening concentration is greater than the screening value, then the exceeding compound is further evaluated for *potential* cancer or non-cancer health impacts on the assumed exposed population.

Further investigation could include, but is not limited to: (a) additional sampling to better estimate a screening concentration; (b) referral for Health Consultation to the PA Department of Health; (c) enhanced investigation of the monitored area for source contribution and impacts; (d) air modeling, and/or (e) refining the population exposure assumptions.

Given the percent data availability for 32KC being below the recommended 85% per quarter and no higher than 69% for three of the eight quarters sampled (see Table 8), using an arithmetic average of the detected values for metals without non-detects (manganese and zinc) as a screening concentration could yield values that are inaccurate and either over- or under-estimating a mean. For instances where incomplete data may potentially bias an estimate of a mean using arithmetic averaging, an Upper Confidence Limit (UCL)¹² of the arithmetic mean can be calculated. As UCL

¹² A UCL is an upper bound probability estimate of a range of values for an estimate of a mean, typically at the 95th percentile, and is denoted as 95UCL. It is a value (concentration) that 95% of the time will be equal to or greater than the arithmetic mean and provides a more suitable screening value for health-based evaluations of ambient air

calculation methods can vary depending on the quality and quantity of the data, a statistical tool (ProUCL¹³) is used to calculate and suggest appropriate UCLs to use on varying data sets.

For compounds where a portion of the collected samples have non-detects, alternative methods for mean estimation and UCL calculation are recommended. In the past, a method of data substitution of values for non-detected data has been used for screening ambient air data. One half or 50% of a compound's analytical reporting or detection limit (1/2RL or 1/2DL) would be substituted for a concentration value when a sample indicates a non-detect. The subsequent data set of detected and substituted non-detect data is then used to estimate a mean by calculating the arithmetic average of the values. While this method is easy and can be done without tedious calculations, research has indicated that the method can significantly underestimate or overestimate a true mean when compared to non-substitution methods.

EPA recommends using the distribution-free Kaplan-Meier (K-M) method for mean estimation on data sets with non-detected data¹⁴. While this method may have been regarded as overly tedious and complicated for non-statisticians, faster computing capability coupled with the development of easier to use and understand statistical packages such as "ProUCL" improve the ability to estimate a more reliable screening concentration over the traditional substitution methods.

For comparison, Table 10 summarizes the minimum and maximum detected 24-hour concentrations, mean concentration estimates by method for the screened compounds (both with and without non-detects) and the screening concentration. As recommended, due to the reduced availability of data, the 95UCL method values were selected as the screening concentration for this analysis.

Table 10 - Summary of Mean Concentration Estimates and Screening Concentrations

Analyte	%ND	Minimum Detect	Maximum Detect	Arith. Mean of Detects	1/2 RL Average	K-M Mean	95UCL	Screening Concentration
Manganese	0%	0.0015	0.031	0.0090	N/A	N/A	0.010	0.010
Zinc	0%	0.0095	0.26	0.039	N/A	N/A	0.056	0.056
Arsenic	35%	0.00067	0.0040	0.0016	0.0012	0.0012	0.0014	0.0014
Lead	57%	0.0045	0.019	0.0073	0.0045	0.0056	0.0060	0.0060
Cadmium	66%	0.00024	0.0017	0.00057	0.00027	0.00033	0.00037	0.00037

data when the collected data may not be systematically representative. By using a 95UCL, one can be certain that 95% of the time the true mean concentration is below the UCL value. It is mathematically based on a calculated mean adjusted depending on the magnitude of the variance of the sample distribution.

¹³ ProUCL is a U.S. EPA-developed and supported software package designed for use in mean estimation, UCL calculation, hypothesis testing and basic trend analysis of environmental data sets. Version 5.0 was used for this report. The software and technical documentation on the methods can be found at <https://www.epa.gov/land-research/proucl-software>.

¹⁴ The K-M method was developed in the late 1950s for use in survival statistics in clinical health studies when monitored patients dropped out, moved or became unavailable for study. Over the years it has grown as a method for estimating descriptive statistics on environmental data sets where non-detected values are present and data might not conform to a known distribution. The ProUCL technical documentation explains the application of the K-M method for the software.

4. Potential Chronic Health Effects Screening

For evaluation of ambient air monitoring data for potential long-term health effects from continued exposure to hazardous metals, the Toxics Monitoring Section uses health-based screening values published by the U.S. EPA, Agency for Toxic Substances and Disease Registry (ATSDR), California Environmental Protection Agency (CalEPA) and others. The values are for lifetime non-cancer inhalation hazard. Screening values for excess lifetime cancer risk are provided for those metals identified as carcinogenic in humans where sufficient data exists to develop risk-based factors.

EPA's Office of Air Quality Planning and Standards (OAQPS) publishes a list of values evaluated from a variety of sources that air quality risk assessors can use for screening of health-based effects.¹⁵ These values are based on respirable particulate (PM₁₀) and not TSP as sampled for Phase II. The values also consider potential effects on sensitive subpopulations (e.g., children).

The Texas Commission on Environmental Quality (TCEQ) Toxicology Division publishes health-based Air Monitoring Comparison Values¹⁶ (AMCVs) for either short-term (1-hour to 24-hour) or long-term (annual) averages of measured ambient air concentrations collected from TCEQ's statewide ambient air monitoring network. These values have been developed specifically to take into consideration differences between health-based values created for use in ambient air toxics exposure modeling for source permitting and real-world ambient data. The single health-based AMCV is the lowest concentration at which either a non-cancer hazard, excess cancer incidence or, in some cases, an objectionable odor could be detected or expected. Observed values greater than the AMCV would be a strong indication of a human health (inhalation hazard or increased cancer risk) or welfare (odor) impact in the monitored area.

Both OAQPS-published values and TCEQ AMCV's for the screened compounds are summarized in Table 11.

¹⁵ Besides use for health-based screening, these values are also used for the permitting analysis of stationary sources subject to air toxic pollutant emissions limitation standards (e.g., MACT and NESHAP programs). The list for chronic exposure values can be found at <https://www.epa.gov/sites/production/files/2014-05/documents/table1.pdf>.

¹⁶ TCEQ AMCVs and supporting documentation can be found at https://www.tceq.texas.gov/assets/public/implementation/tox/monitoring/comparison_values_monitoring.xlsx.

Table 11 - Health Effects Screening Value Summary

Chemical Summary		Non-Cancer Hazard		Cancer Incident Risk		TCEQ Long-Term AMCV (ug/m ³)
Analyte	CAS NO.	Value ug/m ³	SOURCE	IUR 1/(ug/m ³)	SOURCE	
Arsenic compounds	7440-38-2	0.015	CAL REL	0.0043	IRIS	0.067
Cadmium compounds	7440-43-9	0.010	ATSDR MRL	0.0018	IRIS	0.010
Lead compounds	7439-92-1	0.15	EPA NAAQS	0		0.15
Manganese compounds	7439-96-5	0.30	ATSDR MRL	0		0.20
Zinc Compounds	7440-66-6	N/A		N/A		2.0
Notes:						
CAL REL - CalEPA Reference Exposure Level			IRIS - Integrated Risk Information System (EPA)			
ATSDR MRL - Agency for Toxic Substances and Disease Registry - Minimal Risk Level			TCEQ - Texas Commission on Environmental Quality - Toxicology Division			
EPA NAAQS - National Ambient Air Quality Standard						
IUR - Individual Unit Risk		AMCV - Air Monitoring Comparison Value				

Non-Cancer Health Effects Screening

Four of the five metals suitable for screening have non-cancer inhalation health effects screening values for chronic exposure. Potential chronic non-cancer inhalation hazards can be screened from ambient air concentrations by comparing the ratio of the screening concentration to the screening value. If the resulting hazard quotient (HQ) is greater than 1, then a potential chronic inhalation hazard may be present in the monitored area. The TCEQ AMCV is used for zinc screening as EPA, ATSDR and CalEPA have no approved health-based screening value for zinc. Monitored areas where a screening concentration is greater than the AMCV should be evaluated for potential health-based impacts. These calculations and comparisons are presented in Table 12.

Table 12 - Summary of Inhalation Non-Cancer Hazard Screening

Analyte	Screening Value SV (ug/m ³)	Screening Conc. SC (ug/m ³)	HQ (SC/SV)
Arsenic	0.015	0.0014	0.09
Cadmium	0.01	0.00037	0.04
Lead	0.15	0.0060	0.04
Manganese	0.3	0.01	0.03
Zinc	AMCV		SC>AMCV?
	2.0	0.056	No

All screened metals had an HQ below 1.0 and were less than the AMCV. This indicates that the ambient air concentrations of metals observed at site 32K in Phase II were below levels considered unhealthful for chronic exposure for the two-year monitored period.

Cancer Risk Screening

Two of the five metals suitable for screening have a potential risk for a lifetime increase for the incidence of cancer from ambient air. Chronic inhalation exposure to arsenic is known to cause

lung cancer in humans, though mostly through occupational exposure. Cadmium likely causes cancer in the lung, trachea and bronchus in humans and like arsenic, it also is mostly through workplace exposure.

Excess lifetime cancer risk is the probability of an additional incidence of cancer within an exposed population over a lifetime of exposure. It is estimated for each analyte by multiplying the Individual Unit Risk (IUR) by the screening concentration. The sum of the products is the probability of an additional incidence occurring within the exposed population over a lifetime. EPA has established a target lifetime cancer risk from ambient air exposure of between 1-in-1 million people (expressed as 1.0E-06) and 1-in-10,000 people (expressed as 1.0E-04) in an exposed population.

Table 13 summarizes the lifetime cancer risk calculations expressed as a risk probability.

Table 13 - Summary of Lifetime Cancer Risk Screening

Analyte	IUR (1/ug/m ³)	Screening Conc. SC (ug/m ³)	95% UCL Lifetime Cancer Risk (IUR*SC)
Arsenic	0.0043	0.0014	6.0E-06
Cadmium	0.0018	0.00037	6.7E-07
Cumulative Cancer Risk:			6.7E-06

Arsenic in ambient air is the major risk driver of the screened toxic metals. Combined with the smaller risk from Cadmium exposure, the cumulative lifetime cancer risk of 6.7 additional incidents of cancer per one million (6.7E-06) is within the target range of acceptable cancer risk established by the EPA.

5. Health Effects Screening Summary

All screened ambient air toxic metal concentration screening values, using the 95UCL of the two-year screening period mean concentration, were below health-based chronic inhalation hazard and lifetime cancer risk values. For that reason, the observed ambient air concentrations at the monitored location would likely not pose a significant chronic health risk from those metals.

Limitations of Conclusions

The conclusions are only potential health effects for those metals sampled and detected and are only for chronic, long-term exposure. Certainty in the calculation of the screening value is limited by the reduced data availability over the two-year Phase II sampling period; however, this uncertainty has been reduced by using the 95UCL of the 2-year mean concentration of ambient air metals.

This screening assumes that the screening concentration remains the same throughout the lifetime of the exposed population. This assumption is not a prediction of future pollutant levels. Likewise, the screening is only for the two-year sampling period in Phase II and not for the duration of Phase I sampling.

V. Conclusions of Phase I and Phase II Analyses

- The samplers were sufficiently located and operated to collect near-continuous daily ambient air TSP, sulfate and iron concentrations from 1995 through 2013.
- Available meteorological data (wind speed and direction) used to site monitors for the Keystone Cement facility demonstrated that the samplers were in a portion of a predominant downwind vector from the facility.
- TSP concentrations at the site dropped by 28% averaged over the three years following a major conversion at the Keystone Cement facility from a wet to dry Portland cement method in 2009. Concentrations of sulfate and iron in the ambient air dropped by 26% and 11%, respectively, over the same periods.
- No exceedances of applicable NAAQS for particulate matter were observed during either phase of sampling. During Phase I sampling, 13 values of sulfate were observed over a CARB sulfate ambient air quality standard of 25 $\mu\text{g}/\text{m}^3$; however, given that the CARB standard is for the PM_{10} fraction and not TSP as measured at 32K, it is unlikely that health was adversely affected, as a significant portion of TSP is non-respirable.
- In Phase II, 24-hour ambient concentrations of arsenic, cadmium, manganese, and lead were representatively sampled every sixth day in 2014 and 2015 and suitable for health-based toxic metals chronic exposure screening.
- Phase II data availability was less than optimal for Phase II, although by using the 95UCL of the estimated mean concentration of a pollutant as a health-based chronic screening concentration, more reliable and conservative (i.e., more protective) estimates for screening are obtained.
- The final screening concentrations of arsenic, cadmium, manganese and lead were below health-based lifetime screening values, thus indicating that the ambient air in the monitored location does not pose a health risk due to chronic exposure to these metals. Measurable concentrations of total chromium and nickel were found only in 4% and 1% of the samples, respectively.
- For Phase II sampling, all ambient air toxic metal screening concentrations, using the 95UCL of the two-year sampling period mean concentration, were below health-based chronic inhalation hazard and lifetime cancer risk values. For that reason, the observed ambient air concentrations at the monitored location would likely not pose a significant chronic health risk from those metals.