Appendix E

District Additions to the Conceptual Model

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This Appendix provides supplemental discussion of key meteorological, atmospheric and emission issues that affect PM2.5 concentrations. EPA recommends starting the PM2.5 SIP analysis by establishing a conceptual description expressing the factors involved in creating the PM2.5 concentrations found in the nonattainment area. A full conceptual description must include:

- The natural processes of climate and meteorology that cause PM2.5 to accumulate and form additional PM2.5 from precursors
- Atmospheric factors that cause PM2.5 to form or be removed
- Emissions patterns and events that require special consideration

The SJV PM2.5 SIP Modeling Protocol discusses many of the technical issues involved in the conceptual model of PM2.5 for the Valley and references a conceptual model document that was developed from research conducted for the CRPAQS/IMS 95 study to explain the dynamics of both PM10 and PM2.5 events. The discussion in this Appendix was developed by the District to further explain key factors associated with PM2.5 in the San Joaquin Valley that are not fully addressed by the Protocol or the referenced conceptual model document. A variety of special subjects have been assessed by the District to provide a comprehensive review of technical issues associated with PM2.5 concentrations that occur in the Valley. This discussion completes the District conceptual description of PM2.5 as a technical foundation for the PM2.5 SIP.

In addition to use in the development of the SIP, elements of the comprehensive conceptual description are in use on a daily basis to support air quality forecasting. Understanding conditions associated with increases in PM2.5 concentrations provides opportunity to alert the public to conditions that may result in poor air quality and supports decision making for smoke management and residential burning curtailments to prevent air quality deterioration.

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1. Climate and Meteorology

The San Joaquin Valley Air Pollution District (District) analyzed meteorological and air quality data in support of the 2008 PM2.5 Plan. PM2.5 data have been collected since 1999. Currently, there are 11 Federal Reference Method (FRM) PM2.5 air-monitoring sites in the San Joaquin Valley, with at least one monitor in each county except Madera. The cities of Bakersfield and Fresno/Clovis have three sites each due to federal requirements related to their large populations.

1.a Seasonal Differences

PM2.5 is a seasonal pollutant that tends to peak during the fall and winter (late September into February - when nitrate and seasonal carbon sources dominate) compared to the spring and summer when PM2.5 tends to be lower (March into early September with mostly geologic soil and road material, elemental carbon and some secondary carbon aerosol). The 1999 and 2006 PM2.5 measurements from the Fresno 1st Street monitoring site are illustrated in Figure 1. Historically, PM2.5 levels were higher in 1999 compared to 2006. In 1999, the Fresno-1st site experienced several days above the NAAQS, whereas in 2006, only one day was above the 65-microgram standard.

Figure 1 PM2.5 Federal Reference Method (FRM) Measurements for Fresno-1st for 1999 and 2006 The 65 microgram 24-hour average NAAQS is shown on the chart



1.b Meteorological Influences on PM2.5 Concentrations

The PM2.5 concentration can be influenced by a variety of meteorological factors. The District has identified and evaluated several parameters that merit special attention in discussion of the conceptual description of PM2.5 in the Valley: rainfall effects, low wind speed stagnation, reduced mixing height inversion layers, 850mb stability (a regional temperature profile surrogate) and multi-day episode stability. A final review of the 850mb stability parameter shows that the PM2.5 level associated with this parameter is declining as Valley emissions are reduced.

1.b.1 Rainfall Effects

In the warm air mass ahead of an approaching cold front, **concentrations of fine particulate can significantly increase during periods of light rainfall**. Particulate can increase or stay at high levels until the new air mass arrives following the surface frontal passage. During the Bakersfield precipitation event on February 10 and 11, 2005, PM2.5 nearly doubled during a nine-hour period of light rainfall, as shown in Table 1 and Figure 2. As shown in the weather maps provided in Figures 3 and 4, the rain was falling during prefrontal conditions.

Date	Hour	PM2.5	Weather	Temp	RH	WD	WS
		(µg/m ³)		(deg F)	(%)		(mph)
10 Feb 05	20	38	Haze	53	80	CALM	CALM
10 Feb 05	21	37	Fog	52	86	S	5
10 Feb 05	22	40	Fog	53	86	ESE	3
10 Feb 05	23	44	Light Rain/Fog	51	89	NNW	3
11 Feb 05	0	51	Fog	52	89	NNE	3
11 Feb 05	1	54	Light Rain	51	92	W	3
11 Feb 05	2	65	Light Rain	52	86	N	7
11 Feb 05	3	70	Light Rain	53	83	N	3
11 Feb 05	4	58	Light Rain	52	89	N	5
11 Feb 05	5	54	Light Rain	50	94	WNW	7
11 Feb 05	6	52	Light Rain/Fog	50	94	WNW	6
11 Feb 05	7	75	Light Rain/Fog	50	94	WNW	5
11 Feb 05	8	75	Light Rain/Haze	53	83	N	8
11 Feb 05	9	70	Haze	54	80	N	9
11 Feb 05	10	68	Haze	58	75	N	12
11 Feb 05	11	51	Haze	59	67	NNW	9

 Table 1 PM_{2.5} Data from Bakersfield - California Monitoring Station and Weather Data from Bakersfield - Meadows Airport

Temp = Temperature, RH = Relative Humidity, WD = Wind Direction, WS = Wind Speed

Figure 2 PM2.5 During Rain Event in Bakersfield, February 10th and 11th, 2005 Darker shaded bars indicate precipitation reported during the hour



PM2.5 DURING RAIN EVENT

Figure 3 Surface weather map for 4 PM PST (Hour 16) on February 10, 2005 Shading indicates light precipitation







1.b.2 Wind Speed

Stagnant conditions can lead to a buildup of emissions near the emission source area, producing a gradual increase in pollutant concentrations. The longer the stagnant period lasts, the higher the particulate concentrations can rise. An increase in winds causes an end to the stagnant conditions and can transport the polluted air mass downwind, resulting in a lowering of particulate concentrations in the emission source area. Figure 5 illustrates that during stagnant (low wind speed conditions), PM2.5 levels can be high. During the early morning hours of February 14, 2006, winds were less than 3 MPH, resulting in poor dispersion conditions and elevated PM2.5 readings (generally above 50 μ g/m³). After 10 PST, the winds gradually increased and the PM2.5 levels decreased. This plot shows the response of particulates to changes in wind speed.

1.b.3 Mixing Height and Inversion Layers

Vertical mixing height is the elevation in the atmosphere into which emissions will freely mix and disperse. In Figure 6, the base of the inversion layer is equivalent to the mixing height. Vertical mixing is reduced when an inversion layer or temperature inversion is present. A temperature inversion is a deviation from the typical temperature profile in a

well-mixed atmosphere, where temperature decreases with height. Temperature decreases with height in the layer defined by the mixing depth in Figure 6.



Figure 5 Plot of real-time PM2.5 measurements (μg/m³) and wind speed (mph) at Fresno-1st on February 14, 2006

Figure 6 Temperature sounding plot with an inversion layer present



Typically, an inversion is a layer of air warmer than the layer below it as shown in Figure 6. The warmer layer aloft prevents the usual rising airflow that disperses emissions from the ground because the cooler air below the inversion layer does not rise through the warmer air above. As shown in Figure 6, air contaminants accumulate near the ground as emissions continue to be released into the cooler layer near the surface. Temperature inversions play a crucial role in trapping PM2.5 near the San Joaquin Valley's surface. An example of a temperature inversion is from an aircraft measurement from Fresno on October 24, 2007 in Figure 7. The inversion strength was 19 degrees Fahrenheit from the surface up to 1,000 feet. Figure 7 is an example of a surface based inversion. A surface based inversion is a temperature inversion with the base at the earth's surface.



Figure 7 Aircraft temperature measurement from Fresno on October 24, 2007

Statistical analysis has shown that the 850 MB Stability Parameter correlates with PM levels and is an indicator of how much dispersion is occurring in the atmosphere. The District defines the 850 MB Stability Parameter as the inversion located between approximately 5,000 feet (850 MB) and the surface. The 850 MB stability parameter is calculated by taking the 12 Z (4:00 Pacific Local Time) 850 MB temperature at Oakland and subtracting the minimum surface temperature at Fresno (T850 MB (Oakland) – TMIN (Fresno)). A positive stability parameter indicates warmer temperatures with height. A negative stability parameter shows cooler temperatures with height. Figure 8 shows the Fresno-1st 850 MB stability parameter and PM2.5 plot for 2006. PM2.5 is depicted as the solid line, and the stability parameter is shown as the dots at temperature of 75. Figure 8 shows that during periods of strong stability, PM2.5 tends to climb or remain high. A stable atmosphere would be characterized as any positive temperature difference in degrees Fahrenheit. The graph also shows the seasonality of PM2.5 in that during the summer when stability is strong, PM2.5 is low. PM2.5

formation in the atmosphere tends to be more pronounced during the fall and winter months compared to the summer.



Figure 8 Graph showing the Fresno-1st stability parameter and Federal Reference Method (FRM) PM2.5 measurements for 2006

PM2.5 tends to follow the atmospheric dispersion conditions of the San Joaquin Valley. During multiple days of a stable atmosphere, local emissions accumulate under poor dispersion conditions resulting in accumulating PM2.5 concentrations. Preliminary analysis of comparable strength meteorology in 1999 / 2000 and 2005 / 2006 shows that the stronger stability meteorology in recent years is not resulting in the same high levels of PM as in the past. This may be an indicator that District control measures are contributing to the lower levels of PM in the present.

Table 2 illustrates that PM2.5 fluctuates according to local emissions and meteorology. Strong high pressure built over the region on February 5, 2006 trapping PM2.5 near the surface. PM2.5 levels continued to climb under stagnant dispersion conditions until on February 13 when the Fresno-1st monitor recorded 71 μ g/m³.

Date	PM2.5 Measurement
2/4/2006	22
2/5/2006	26
2/6/2006	30
2/7/2006	26
2/8/2006	38
2/9/2006	49
2/10/2006	56
2/11/2006	51
2/12/2006	56
2/13/2006	71
2/14/2006	34
2/15/2006	8

Table 2: Federal Reference Method (FRM) PM2.5 (μg/m³) Measurements at Fresno-1st Monitoring Site

PM2.5 and the 850 MB stability parameter were analyzed from 1999 to 2006 during the peak season (November 10 through January 15), Figure 9. This analysis was conducted to determine if meteorology or District control measures played a role in the current years' (2005 / 2006) PM2.5 levels. In Figure 9, stability increases from left to right, whereas PM increases from bottom to the top. In 1999 and 2000, high stability days tended to result in elevated PM2.5 levels well above the NAAQS. Recently in 2005 and 2006, high stability days resulted in lower levels of PM2.5, typically near or below the NAAQS.

Figure 9 PM2.5 measurements at Fresno-1st related to Oakland 850 MB –Fresno temperature stability parameter. High stability days (850 MB stability parameter is greater than 0) resulted in high PM2.5 measurements from 1999 through 2001. Comparable high stability days since 2003 have not caused high PM2.5 measurements.



1.c Significant Changes in AQI Due to Meteorological Events

Very large changes in the Air Quality Index (AQI) and PM2.5 concentration can occur over a short time period during the PM2.5 season. Figure 10 presents a plot of the AQI in Tulare County (Visalia) during the 2005-2006 PM2.5 season. An AQI improvement of 134 occurred from the 25th to the 26th of November 2005. Stagnant conditions were in place over the San Joaquin Valley Air Basin on the morning of November 25, 2005 as shown in the Figure 11 weather maps. A vigorous weather front passed over the region during the afternoon and evening of November 25. PM2.5 concentration decreased rapidly under the influence of a new clean air mass and moderate NW winds. The tightly packed isobars on the weather maps are an indication of a strong pressure gradient and the wind flags indicate moderate winds. The Visalia PM2.5 concentration decreased rapidly around midnight as shown in Table 3 and Figure 12. Figure 12 displays how the decrease in the PM2.5 concentration progresses from northwest to southeast in the San Joaquin Valley Air Basin, coinciding with the front's progression from northwest to southeast across the Valley. The decrease in PM2.5 first occurred in Stockton in the north valley. South Valley stations at Corcoran and Visalia recorded the decrease in PM2.5 later in the day.



Figure 10 AQI in Tulare County (Visalia) During the 2005-2006 PM 2.5 Season

able 3 PM2.5 concentration in Visalia, November 25th ar	1d 26 th , 200؛	5
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Date / Hour	Visalia PM2.5	Visalia Municipal Airport
	(µg/m3)	Wind Direction and Speed (mph)
25 November 2005 Hour 13	89	ESE 9
25 November 2005 Hour 14	91	NE 6
25 November 2005 Hour 15	85	N 5
25 November 2005 Hour 16	68	N 5
25 November 2005 Hour 17	72	N 3
25 November 2005 Hour 18	78	WNW 10
25 November 2005 Hour 19	88	WNW 12
25 November 2005 Hour 20	79	NW 10
25 November 2005 Hour 21	76	NNE 5
25 November 2005 Hour 22	63	NNW 9
25 November 2005 Hour 23	39	NNW 10
26 November 2005 Hour 0	27	NW 10
26 November 2005 Hour 1	22	NNW 14
26 November 2005 Hour 2	11	NNW 7
26 November 2005 Hour 3	6	NW 8
26 November 2005 Hour 4	2	WNW 5
26 November 2005 Hour 5	3	NW 7



Figure 11 Surface Weather Maps for 4 AM (12Z), November 25th and 26th, 2005



Figure 12 PM2.5 Concentration, November 25th and 26th, 2005

The southern San Joaquin Valley Air Basin experiences significant decreases or increases in AQI followed by relatively constant AQI, as may be seen in Kern County data presented in Figure 13. The significant decreases can occur in one day. The significant increases tend to take several days to build. The northern San Joaquin Valley Air Basin experiences more frequent significant increases and decreases in AQI, as may be seen in San Joaquin County and Stanislaus County data presented in Figure 14 and 15. This is likely due to more frequent frontal passages in the northern San Joaquin Valley. In the south, a front that may have influenced the north valley may dissipate before reaching the southern San Joaquin Valley, resulting more potential for longer periods of stagnation than the north Valley.



Figure 13 AQI in Kern County (Bakersfield)









The most significant decreases in PM2.5 tend to occur when there is an air mass change associated with a vigorous frontal passage, typically in fall and winter. It appears that the mechanism that lowers the PM2.5 concentration is an air mass change and not necessarily by collision - coalescence or 'washout' by falling precipitation. As discussed in section 1.b.1 of this Appendix, precipitation can sometimes be associated with a significant increase in PM2.5 in the San Joaquin Valley.

There is scientific support for why collision - coalescence may not be the primary mechanism for PM2.5 removal. Collisions between water drops and particles are likely a function of gravitational, electrical and aerodynamic forces. As a water drop falls in a cloud, it will collide with a very small amount of the particles in its path because aerodynamic forces tend to move the particles to the side, out of the drop path. If a collision of a water drop and PM2.5 particle occurs, this collision does not guarantee coalescence and removal of PM2.5. Rogers (1976) states that studies of aerodynamic forces in cloud drops have indicated that very small drops cannot readily be made to collide. A small drop falling though a cloud of smaller droplets will collide with a very small percentage of drops in the drop path as long as the drop radius is less than 18µm (Rogers, 1976). The drops may bounce away from each other; they may coalesce and remain intact; and they may coalesce briefly and separate. The process is a function of water drop size, particle size, drop trajectories and external forces such as electrical charges. Therefore, some precipitation processes may not be sufficient to cause droplet capture of PM2.5. Air mass exchange appears to be a more effective PM2.5 removal process in the Valley.

Very large AQI and PM2.5 concentration changes can also occur over a short time period during relatively fair weather conditions. An AQI improvement of 89 occurred from the 15th to the 16th of November 2007 in the north San Joaquin Valley (112 AQI to 23 AQI in Modesto) during a period of benign meteorological conditions. An AQI improvement of 67 occurred in the south San Joaquin Valley (in Visalia from the 16th to the 17th of November 2007). The improvement in AQI was likely due to a subtle improvement in dispersion conditions, as meteorological instrumentation recorded small changes in parameters. Water vapor satellite images indicated a very weak frontal passage occurred in zonal flow, resulting in a partial air mass change. The weak front was not evident in the surface weather maps. Physical evidence of the frontal passage was registered by an increase in the mixing height, a slight increase in surface winds and some high and middle clouds.

1.c.1 References for Significant Changes in AQI Due to Meteorological Events

- Mesowest website, http://www.met.utah.edu/mesowest/, Archived weather data
- Rogers, R. R. (1976), A short course in cloud physics, Pergamon Press
- UNISYS website, http://weather.unisys.com/, Archived weather maps

2. Physical and Chemical Processes

A variety of physical and chemical processes occur in the atmosphere causing PM2.5 to form or to be removed, including fog processing, dry deposition and nitrate production. Due to historical episodes fog relationship with particulate has been a subject for extensive research. California and the San Joaquin Valley experience high levels of nitrate particulate that is not a common pattern for other states. The District is evaluating nitrate patterns and has identified information of potential importance for attainment planning. Carbon is another area requiring major continuing evaluation. Elemental and organic carbon are directly emitted, transformed by chemical processes from gases to particles, and are sampled in error as particles by absorption of gases on sampler filters (sampler artifact) making it difficult to assess.

Some factors have an effect but are considered as either less critical or not well quantified for current use in our decision-making processes. Temperature and relative humidity affect properties of aerosols but have not been determined to be critical factors. Dry deposition of particulates has not been extensively studied. Particle deposition and aerosol re-evaporation flux are not well quantified and are not included in District forecasting and modeling.

2.a PM2.5 Formation and Removal During Fog Episodes

The District has examined fog effects on particulate pollution to understand the role of fog on Valley air quality. Scientific investigation of the effect of fog is warranted based on historical events that demonstrate a lethal potential for severe fog episodes accompanied by PM2.5 high levels of carbon emissions. Probably the most famous episode occurred in London, England from 5 to 9 December 1952, resulting in the deaths of thousands. In the London episode, a cold fog developed resulting in the burning of more coal than usual. The polluted air mass was trapped under a strong temperature inversion layer, which acted like a lid on the air mass with coal soot accumulating to lethal levels. Research was conducted in the San Joaquin Valley to explore the consequences of severe fog events in the Valley and determine the severity of PM2.5 during fog episodes.

While much has been learned about fog interactions with key inorganic aerosol species (e.g., ammonium sulfate and nitrate), it is only in recent years that investigators have begun examining interactions between fogs and carbonaceous aerosols and volatile organic compounds. The District has examined fog effects on particulate pollution through integrated projects included in the major field study the California Regional Particulate (PM10/PM2.5) Air Quality Study (CRPAQS) and through special technical studies and dedicated fog research projects. Special projects related to fog were conducted in a preliminary field assessment for CRPAQS known as the 1995 Integrated Monitoring Study (IMS95) and in subsequent projects to assess fog and fog chemistry. These studies examined PM2.5 formation and removal during fog episodes. Significant

CRPAQS findings concerning fog are presented in the following pages and include excerpts from a CRPAQS report written by J. Collett, Jr. et al (2005).

2.a.1 Fog Climatology

During the winter, persistent high pressure over the Great Basin creates a strong subsidence inversion over California's Central Valley, with a base typically a few hundred meters off the Valley floor and below the surrounding mountain ridges (Holets et al., 1981). With the help of the mountains, this strong inversion forms a lid over the air basin, trapping cool, moist air within the valley. Subsiding air results in clear skies, providing excellent conditions for strong radiative cooling at night and, if sufficient moisture is present, the formation of dense, widespread radiation fogs.

2.a.2 Fog Interactions with Gases and Particles, Particle Formation and Removal

Fog is defined as water droplets suspended near the earth's surface that reduce visibility below 1 km (Huschke, 1959). Fog differs from a cloud in that the base of fog is at the earth's surface, while clouds are above the surface. Fog water droplets are tiny, ranging in size from several micrometers to several tens of micrometers. This compares to the diameter of one PM2.5 particle for the smaller dimension, ranging to several times the diameter of a PM10 particle for the largest fog particles. These drops form by water vapor condensation onto aerosol nuclei known as cloud condensation nuclei (CCN). Water-soluble constituents in the CCN determine the initial composition of the fog.

Particles that become fog droplets by absorbing water vapor are subsequently removed by gravitational settling. Fog episodes cause a net removal of PM by wet deposition. However, fog also provides an aqueous medium for formation of sulfate particulate. Fogs have two important, competing effects on aerosol populations:

- New aerosol mass formation through gas scavenging and chemical reaction in the droplets leading to non-volatile species (e.g. conversion of SO₂ to sulfate) that remain in the particle phase after droplet evaporation
- Aerosol scavenging followed by deposition through droplet settling and/or impaction

The relative importance of these two processes depends on the environment in which the fog forms and can include meteorological conditions, number and composition of aerosol particles, and gas phase chemical composition. The net effect of a fog episode depends on the pre-fog concentrations of particulate precursors and availability of oxidants that are involved in the chemistry of particle formation, and the duration of the fog event. The net effect of a fog on atmospheric aerosol concentrations may change during a fog event; oxidation could be more important at the beginning of the fog event when reactant concentrations are higher, while deposition rates may increase over time with the growth of fog droplets.

Previous studies of sulfur oxidation in San Joaquin Valley fogs have shown that dissolved sulfur dioxide can react rapidly, either being oxidized to sulfate or reacting with carbonyl compounds to form hydroxyalkylsulfonic acids. Sulfate is both produced and removed by fog. The end result of a fog event may be either and increase or decrease in the monitored levels of PM2.5 sulfate. Sulfate particulate is removed by wet deposition during fog; however, fog droplets provide the aqueous medium for the production of sulfates, and the production of additional sulfate particulate by chemical reactions within the fog may over-balance the wet removal and actually increase the amount of sulfate PM2.5 in the atmosphere. Whether sulfate formation in fog exceeds sulfate removal by fog settling will vary from site to site, depending on the duration of event and concentrations of available reactants to form additional sulfate particulate matter. Variations in fog drop composition with size are known to influence the rates of chemical reactions as well (e.g., Reilly et al., 2001).

Fog can process particles and gases as shown in Figure 16. Growth of droplets formed with aerosol CCN lead to removal of particles and gases from the air mass, which can lead to a decrease in PM2.5 concentration.



Fog droplet composition is also influenced by dissolution of soluble gases and aqueous phase chemical reactions as shown in Figure 17. The competition between removal and production processes leads to either an increase or decrease in PM2.5 concentration. These processes vary with fog characteristics.



Figure 17 Fog Droplet Particle and Gas Scavenging Processes SIMPLIFIED CHEMISTRY OF A FOG DROP

Deposition due to fog drop sedimentation or impaction has been known to be an important removal process for atmospheric pollutants for a long time (Waldman, 1986). Some studies have tried to assess the deposition fluxes by fog in the San Joaquin Valley by modeling (Lillis et al., 1999) or by measurements (Collett et al., 2001). However, there are relatively few measurements for atmospheric removal of fog solutes by drop deposition or how drop-size dependent fog composition affects removal rates for various chemical species.

While much is now known about the inorganic composition of San Joaquin Valley fog water, little is known about the scavenging and removal of carbonaceous aerosol by these fogs. Despite measurement of high total organic carbon (TOC) concentrations (e.g., during IMS95), the composition of the organic species making up this TOC is largely unknown in the San Joaquin Valley and elsewhere. Although the expected net effect of San Joaquin Valley fog episodes is reduced atmospheric loadings of carbonaceous aerosol, the magnitude of this removal was unknown prior to CRPAQS. Secondary Organic Aerosol (SOA) formation (formation of organic carbon particulate from photochemical oxidation reactions of precursor gases) is reduced by decreased photochemical activity in circumstances where sunlight is reduced by fog or cloud cover. Clouds and fog reduce the photochemical production of secondary organic aerosols by a factor of two or three from the maximum clear sky conditions because of

reduced availability of photons. The thermodynamic interactions between fog droplets and SOA are unknown.

2.a.3 Fog Research Results

In order to improve the understanding of the role fogs play in influencing aerosol concentrations in California's Central Valley, Colorado State University (Collett et al, 2005) made measurements of the chemical and physical properties of fogs at several valley locations during the CRPAQS winter intensive. These observations subsequently formed the basis for a data analysis effort aimed at better characterizing fog composition and interactions between CRPAQS fogs and pollution levels, especially fine particles. The fog measurement campaign and subsequent data analysis phase produced a number of significant findings. These include the following major observations and conclusions made by J.Collett, Jr. et al (2005):

- Several fog episodes were characterized during CRPAQS. Most of the fog episodes were relatively shallow and featured very large droplets. Rarely did the top of the fog layer reach even 100 meters, in contrast to much deeper fogs observed during IMS95 and other studies in the San Joaquin Valley. The very large drops formed during CRPAQS probably result from strong radiative cooling directly from the drops themselves, through the shallow fog layer, driving rapid condensational growth. These large drops settle from the fog relatively quickly, enhancing the ability of the fogs to rapidly remove scavenged airborne particulate matter.
- The chemical composition of the fogs during CRPAQS was dominated by nitrogen species, with important contributions also from organic compounds and sulfate. Ammonium and nitrate were the most abundant individual compounds; nitrite and sulfate were also found to be present at significant concentrations, as were several organic compounds, including formate, acetate, and formaldehyde. Abundant gas phase ammonia absorbed by fog drops helps keep the fog pH relatively high, with pH values typically well above 6. Material present in fog drops is derived from a combination of aerosol particle scavenging, gas absorption, and aqueous phase reactions.
- Comparison of Bakersfield fog composition measurements in CRPAQS and other recent San Joaquin Valley fog studies with measurements made in the 1980's reveals a statistically significant decrease in fog concentrations of sulfate and an increase in fog pH. These changes are consistent with intervening declines in SO₂ emissions in the southern San Joaquin Valley, which would translate into less production of sulfate and greater availability of ammonia to raise fog pH.
- Comparison of fog composition during CRPAQS at Bakersfield, Angiola, and Helm reveals that Angiola and Helm, both rural sites, have generally similar

compositions. Urban Bakersfield fog contained greater concentrations of sulfate and nitrite.

- The high pH droplets present in CRPAQS fogs make them effective atmospheric reactors for dissolved sulfur dioxide. Both oxidation to sulfate and reaction with dissolved formaldehyde to produce hydroxymethanesulfonic acid (HMS) are important reaction pathways. Numerical simulations using a single drop fog chemistry model reveal the importance of considering effects of mass transport limitations on aqueous sulfur chemistry in the large fog drops observed in CRPAQS fogs.
- Preferential enrichment of major ion species in small fog drops reduces most species' deposition velocities below the deposition velocity for fog water. Nitrite, which was often enriched in large fog drops, exhibited deposition velocities higher than fog water. Accounting for drop size-dependent species concentrations is essential to accurately modeling rates of pollutant deposition in San Joaquin Valley fogs.
- Previous studies have documented the important role San Joaquin Valley fogs play in cleansing the atmosphere via particle scavenging followed by drop deposition. The importance of this mechanism was again observed during CRPAQS, with fog deposition fluxes capable of reducing boundary layer fine particle concentrations of major species (e.g., nitrate and ammonium) at a rate on the order of 1 µg/m³ per hour. The fogs are also effective at scavenging and removing sulfate, but this removal is often offset by similar rates of sulfate production via aqueous phase oxidation of sulfur dioxide. Assuming a 100 m deep fog, Collett et al found that concentrations typically decrease as follows:
 - SULFATE ~ 0.5 μ g/m³ per hour
 - NITRATE ~ 1 μ g/m³ per hour
 - AMMONIUM $\sim 1 \, \mu g/m^3$ per hour
 - TOTAL ORGANIC CARBON ~ 0.7 μ gC/m³ per hour
- Results obtained during CRPAQS also indicate the important role that San Joaquin Valley fogs play in scavenging and deposition of organic carbon. San Joaquin Valley fogs contain a rich mix of organic compounds, with major constituents including formaldehyde, formate, and acetate. Many larger organic molecules are also observed, including both polar and non-polar compounds. Approximately 25% of the fog organic carbon, on average, is present as undissolved, suspended material in the droplets. As much as half of the fog organic matter may be comprised of high molecular weight compounds, with molecular masses exceeding several hundred Daltons. Future studies are needed to better characterize this high molecular weight material and determine whether it comes mainly from aerosol scavenging or is produced by aqueous phase reactions of lower molecular weight precursors.

- The wide variety of organic compounds observed in the fogs indicates that carbonaceous aerosol particles from many source types undergo active fog processing. Observations of fog scavenging of organic and elemental carbon also indicate the importance of fog processing of carbonaceous aerosol species. Organic carbon (OC) was observed to be more actively processed by CRPAQS fogs than elemental carbon. Differences were also observed between the efficiency of CRPAQS fogs in scavenging different types of fine particle organic carbon. Wood smoke particles were scavenged more efficiently and particles containing polycyclic aromatic hydrocarbons (PAH) less efficiently than fine particle OC as a whole. CRPAQS fogs were also observed to efficiently scavenge and remove the pesticide Diazinon. Together, these observations suggest that fogs may be more effective cleansing the boundary layer of OC from some source types than others, a topic deserving further attention in future studies.
- Deposition fluxes of organic carbon in fog water were significant, yielding atmospheric removal rates on the order of 66 µgC/m² hr. Because some of the fog burden of organic carbon is comprised of soluble, volatile organic compounds (VOC), a portion of the deposited carbon may be released back to the atmosphere when deposited fog water evaporates following a fog episode.
- Comparisons of fine particle organic carbon concentrations before and after fog episodes also suggests that aqueous phase reactions of dissolved VOC's may be important in producing new, secondary organic aerosol matter. Much more work is needed to examine this hypothesis in future investigations.
- Measurements of the stability of carbonyl compounds in actual fog water reveal the importance of stabilizing these species as soon as possible following sample collection. Analysis of field-stabilized samples collected in Fresno fog episodes after CRPAQS reveal significant contributions of several carbonyl and dicarbonyl compounds to San Joaquin Valley fog organic carbon burdens. Glyoxal and methylglyoxal, in particular, were observed at substantial concentrations nearly equivalent to formaldehyde.
- The Carnegie Mellon University fog model, a 14-section size-resolved fog model with explicit fog microphysics, was used to simulate a CRPAQS fog episode and was able to predict the liquid water evolution, bulk aqueous-phase concentration measurements, drop size-resolved trends, and deposition fluxes for a number of species in close agreement with observed values. The predicted evolution of the distribution of aerosol species like sulfate, nitrate, ammonium, and chloride indicated significant differences over the course of fog processing. Different sizedependent processes controlled the size-distribution development at different stages of fog processing. Early in processing, the dissolution of soluble gases and rapid aqueous phase reactions control the development of the size distribution. At later times, the effects of deposition begin to dominate, and push

the peak of the distribution towards smaller diameters. Model predictions of the evolution of most key species approached bulk aqueous-phase concentration measurements. During the early stages of the fog, the behavior of species originating partially in the gas phase (NO_3^- , $SO_4^{2^-}$) was heavily influenced by dissolution into the droplets and rapid aqueous phase reactions. Following these initial peaks in aqueous phase concentrations, deposition began to dominate, and the species were gradually depleted from the fog layer.

2.a.4 Fog References

- Collett, J.L.J., Herckes P., Chang H., and Lee T., Pandis, S.N., and Fahey K. (2005) Examination of the Influence of California Regional Particulate Air Quality Study Fog Episodes on Aerosol Formation and Removal, *Submitted to: San Joaquin Valleywide Air Pollution Study Agency, January 13, 2005*
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2.b Dry Deposition

Fine particles are small enough that the random forces from collisions with gas molecules largely overcome gravitational forces. Fine particles tend to follow air streams and are typically not removed by impaction. As a result, they are not easily traced back to their individual sources.

Fine particles formed from accumulation processes are significantly larger than gas molecules and their diffusion velocity is low. These particles have very long half-lives in

the atmosphere, travel long distances, and tend to be more uniformly distributed over large geographic areas than coarse-mode particles. Removal by dry deposition is inefficient since they do not readily diffuse through the boundary layer of still air next to surfaces. The atmospheric half-life of fine accumulation particles with respect to dry deposition is on the order of weeks; however, removal occurs when the particles absorb water, grow into cloud droplets, grow further to raindrops, and fall out as rain. This process reduces the typical atmospheric half-life to a few days.

Ultrafine or nuclei-mode particles, formed by nucleation of low saturation vapor pressure substances, tend to exist as disaggregated individual particles for very short periods of time (less than minutes) in the ambient atmosphere and tend to age rapidly into larger accumulation particles that may be dispersed more widely over long distances.

Secondary fine particles are formed by atmospheric transformation of gases to particles. Atmospheric transformation can take place locally during stagnations or during transport over long distances. Fine particles have very low dry deposition velocities, which contribute to their uniformity throughout the air mass. Aerosol effects on visibility and climate, through light scattering and changes in cloud microphysics, primarily arise from fine particles.

Nitrates and carbon are the two largest components of PM2.5 and their atmospheric behavior is entirely different.

"High concentrations of PM organic carbon (OC) were spatially limited to core urban sites while high concentrations of PM ammonium nitrate were regionally distributed throughout the SJV. Concentrations of PM and its precursors were typically lower at the elevated sites surrounding the SJV than at monitoring sites located on the SJV floor....At distances more than 50 km from the urban areas, OM concentrations typically declined by a factor of three or more. Emissions of OM at the urban core are either not rapidly transported to the rural sites or are diluted too much to substantially impact rural sites. Concentrations of OM at elevated sites were comparable to concentrations at rural sites on the Valley floor....Overall, these spatial patterns of OM suggest that the impact of emissions was largely confined to the local area and OM concentrations were unevenly distributed over the duration of the episode.

The contrast in spatial variability between the ammonium nitrate and OM components of PM2.5 in the SJV winter episodes provides information on the spatial extent of the production of ammonium nitrate. PM2.5 OM and ammonium nitrate are both subject to the same meteorological transport conditions, yet ammonium nitrate concentrations are relatively homogeneous and OM concentrations are much higher in the urban source areas. In addition, OM and ammonium nitrate components are expected to have the majority of their mass in a similar size fraction (PM0.1 to PM1) (Lighty et al., 2000; Hughes et al., 1999; Bench et al.,

2002) and, therefore, the rates of removal should be approximately the same. In summary, the likely explanation for the difference in spatial variability is the spatial distribution of the emissions or precursors. Primary OM emissions occur predominantly from mobile sources and wood smoke located in urban areas. The formation of ammonium nitrate from NOx precursors (Lurmann et al., 2004) must occur throughout the SJV to account for its spatial homogeneity.

High concentrations of PM organic carbon were spatially limited to core urban sites while high concentrations of PM ammonium nitrate were regionally distributed throughout the SJV. The regional homogeneity of ammonium nitrate concentrations coupled with the stagnant wind conditions provides evidence that production of ammonium nitrate occurs at similar rates throughout the valley. In contrast, the OC component of PM indicates that production rates were much higher in the urban areas than at rural sites." (reference citation follows in 2.b.1)

2.b.1 Dry Deposition Reference

 Background and Boundary Conditions for Particulate Matter and Precursors in the San Joaquin Valley in Winter, Technical Memorandum STI-902325-2779-TM, Michael C. McCarthy, Hilary R. Hafner, Steven G. Brown, Fredrick W. Lurmann, Paul T. Roberts, Sonoma Technology, Inc., 1360 Redwood Way, Suite C, Petaluma, CA 94954-1169, July 29, 2005

2.c Nitrate Events

Nitrate particulates are generally low in summer and fall, partially due to breakdown of the particles back into gaseous precursors. Photochemical reactions reduce nitrates; therefore, overcast conditions support increase in nitrate production. Valley summers have infrequent cloud cover. Smoke from fires whether aloft or at the surface can reduce photochemical energy penetration to the surface allowing more particle formation. As with all other particulates, accumulation to high concentrations is also dependent on stagnation events to allow the material to accumulate.

2.c.1 Relationship Between Carbon and Hydroxyl Radicals and Nitrate PM2.5

Nitrate particulate levels in the San Joaquin Valley declined sharply after the 2000-2001 winter. It is difficult to isolate meteorological variations from other factors to determine a strong cause and effect relationship. However, the most pronounced change (during the time of year that the nitrate levels increase) was the introduction of different smoke management practices including the voluntary curtailment (commencing in 2001) and mandatory curtailment (commencing in 2003) of residential wood combustion when air quality conditions were forecast to be unfavorable.

Since the primary emissions from residential wood combustion are carbon and VOC, the link between reduced burning and reduced nitrate particulate levels is not automatically evident. EPA has established that hydroxyl radicals, released by chemical reaction from organic VOC, are the primary mechanism for much of the atmospheric chemistry for both ozone and particulates. The role of hydroxyl radicals in ozone and particle formation is a key concept in EPA's "One Atmosphere" technical evaluation. Nitrate particulates require hydroxyl radicals to form particulate. This establishes the possible effect of receiving multiple benefits from reducing carbon and organic carbon emissions, in that not only will carbon be reduced but also secondary particulate formation including nitrates and secondary organic aerosol (SOA).

Additional evidence supports the relationship between carbon and nitrate particulate formation. A sharp increase in nitrate appears to occur during periods when the air shed is being significantly impacted by wildland fire smoke. During several wildfires that occurred during the summer and fall, when nitrate is usually at very low concentrations, elevated levels of nitrates occurred on a daily basis. This conveys that either the fires produce much more nitrate than expected and transport it to distant areas or that the carbon and hydroxyl radicals promote nitrate formation in the local area. Either circumstance connects fires to nitrates. The carbon in the smoke plume provides carbon nuclei and/or organic carbon radicals that can initiate nitrate particle formation. The District has conducted preliminary evaluation of two case studies that provide supporting information. Further evaluation of these events and other similar events may be warranted to establish a confirmed finding.

Figure 18 Daily average nitrate and maximum hourly nitrate by day at Fresno -First Street monitoring station affected by Day Fire



Nitrate Concentrations - Fresno - First Street

Figure 18 shows high nitrate concentrations during a period of smoke production from large wildland fires. Typical nitrate levels at this time of year (September and early October) are less than ten micrograms.

Figure 19 is a smoke plume simulation using the CANSAC MM5 meteorological model. At the time of this analysis, 10:00 PST on October 10, 2006, the smoke plume was projected to be over the Fresno area. The CANSAC simulation has not been confirmed for accurate fire inputs for this simulation. The value of assessing this type of event is that the path and origin of emissions provide a more direct linkage to a fire event than when the emissions path crosses through large cities. Events of this type do not occur frequently and events that contain emissions mixed from many source areas are more difficult to resolve into cause and effect relationships.

Figure 19 Wildfire smoke simulation for October 10, 2006, at 10:00 PST, displaying peak particulate concentration over the Fresno area



Prescribed Fire & Wildfire Simulation

MM5 Forecast: 2006100900 PM2.5 (NAAQS = 65 micrograms/m^3, 24hr avg)

In Figure 20, the observed increase in nitrates occurred in the hours immediately after the projected arrival of the smoke plume in Fresno. This indicates that the smoke plume itself was not high in formed nitrates, but provided carbon nuclei and/or organic carbon radicals that initiated nitrate particle formation. After departure of the smoke plume, nitrate levels rapidly returned to ambient levels that were measured before the arrival of the smoke plume.



Figure 20 Hourly nitrate (ug/m3) at Fresno - First Street monitoring station, displaying increase in nitrate on October 10, 2006, in the hours following arrival of the smoke plume

The case studies, and unexpected decline in nitrates after establishing stronger burning restrictions, establish a strong likelihood that burning controls provide more than a carbon benefit for the District. Understanding and confirming this relationship through future research and technical evaluation is valuable for effective PM2.5 planning.

3. Emissions Related Issues

High levels of PM2.5 may occur anytime throughout the year if influenced by unusual local or regional emission activity. Wildland fires and 4th of July firework activity have been noted as causing PM2.5 levels to be higher than what is typically experienced during the summer and fall. Day of the week activity variations and holidays during the fall and winter have the potential to cause higher PM2.5 levels due to residential wood burning.

3.a Day of week and Holiday Effect

- **Day Of The Week** Use of fireplaces tends to increase on cold winter weekend nights. PM2.5 can increase on Friday, Saturday and Sunday when residential wood combustion may be higher than during the workweek.
- **Holiday** On winter holidays, residential wood combustion from fireplaces can substantially increase, resulting in a significant increase in the particulate matter concentrations. Days around Thanksgiving, Christmas and New Year's Day tend to have higher PM2.5 concentrations.
- 4th of July PM 2.5 concentrations increase significantly during the evening of July 4 and continue into the early morning hours of July 5 due to fireworks emissions. This effect is especially pronounced during periods of stagnant weather conditions. On July 4, 2007, PM2.5 increased to very high concentrations during the time of day when fireworks were used, as shown in Figure 21. This coincided with a period of stagnant conditions, which resulted in PM2.5 concentrations remaining high into the early morning hours of July 5. Fireworks emissions are categorized as an Exceptional Event by EPA policy, but the District is proposing a feasibility study to investigate the possibility for controls on fireworks (see Chapter 6).



Figure 21 Fireworks Emissions Effect on PM2.5, July 4th and 5th, 2007 Data in this plot is preliminary and subject to revision

3.b Wildland Fires

Wildland fires can have a significant effect on particulate air pollution. Wildland fires can be ignited by human activity and by lightning. These fires are either suppressed (put out) or managed (allowed to burn) as a controlled burn. Particulate from wildland fires can increase surface PM 2.5 concentrations. Figure 22 shows smoke from the Day Fire being transported to the San Joaquin Valley. It should be noted that large wildfires may release particulates in amounts that dwarf the District daily emission inventory, causing impacts that cannot be fully remediated by District action to curtail local emissions. The impact of a wildfire will depend on whether the smoke passes aloft over the District or mixes into the surface air. Wildfires release carbon, NOx and VOC's. Some of the VOC's that are emitted by wildfires are classified as air toxics.



Figure 22 Smoke from the Day Fire Transported to the San Joaquin Valley

3.c Agricultural and Prescribed Burn Management

The District's agricultural, hazard reduction, and prescribed burn programs permit, regulate, and coordinate the use of burning in the San Joaquin Valley air basin to minimize smoke impacts on the public. The goal of these smoke management programs is to minimize air quality impacts by allowing agriculture, hazard reduction, and prescribe burning to occur on days when dispersion is adequate enough to carry emissions away from sensitive receptors. The District has developed a state of the art Smoke Management System (SMS) that allows the District to permit agricultural burning in some areas of the District and not allow agricultural burning in others, based on dispersion conditions and air quality. The SMS reduces smoke impacts by preventing burning from occurring in a concentrated area or in locations that are forecast to have poor air quality, while allowing agricultural burning in areas that have adequate dispersion and good air quality. The District manages the burn programs daily to determine if the potential emissions will impact the air guality in the air basin. On days when the air quality is predicted to be marginal or poor, the District severely restricts, or does not allow, agricultural, hazard reduction, and prescribed burning to occur. When air quality conditions improve, the District allows open burning as dispersion conditions and air quality dictate.

Applicable District rules regulating burning activity: **Rule 4103 Open Burning - Agriculture Rule 4106 Prescribed Burning and Hazard Reduction Burning**

3.d Residential Wood Burning Program

The purpose of the District's residential wood burning program is to protect the public by limiting the emission of particulate matter from wood burning fireplaces and wood burning heaters when air quality conditions are unfavorable for dispersion of the emissions. The District discourages the public from burning when the Air Quality Index (AQI) is greater than 100. An episodic curtailment for a geographic region is declared whenever an Air Quality Index value of 150 or greater is predicted for the geographic region (county). An episodic wood burning curtailment may be issued during the months of November through February. Operation of wood burning fireplaces and wood burning heaters is prohibited in the specified region when the District declares a curtailment, except for certain exemptions (e. g. device is sole source of home heat).

Applicable District rule regulating burning activity:

Rule 4901 Wood Burning Fireplaces and Wood Burning Heaters

3.e Recent Particulate Research Studies

Evaluation of Rule 4901 This study (Lighthall, 2007) is designed to provide the District with a comprehensive evaluation of Rule 4901, which regulates residential wood combustion. Rule 4901 set in place an AQI-triggered ban on wood burning in areas of the San Joaquin Valley that have natural gas service and are below 3,000 ft elevation. In particular, the study will assess the contribution of the Rule to reduced PM2.5 emissions in urban areas of the region and to corresponding reductions in health impacts (mortality and morbidity). In addition, to contributing to the larger body of knowledge about the health effects of PM2.5 exposure, the study will provide the District staff and Board with a solid scientific basis for determining whether additional restrictions in wood burning will be justified.

PM Emission Factors and Inventories from Leaf Blowers The District funded the University of California at Riverside - College of Engineering Center for Environmental Technology (CE-CERT) to design a study and perform measurements and data analysis to determine particulate matter emissions from leaf blowers and to obtain a PM emission inventory from their operation in the District. The following is an excerpt from the executive summary of the final report (Fitz, 2006).

The approach used to measure emissions from leaf blowers and alternative devices (vacuums, rakes, and brooms) was to operate the devices over a measured area in a tent-like enclosure. In this enclosure, the leaf blower (or other device) could be used in a normal manner while allowing the PM emissions to be confined for quantification. PM concentrations were measured with real time sensors. Measurements were made for total suspended particulate matter (TSP), particulate matter (PM) with an aerodynamic diameter less than ten microns (PM10) and particulate matter with an aerodynamic diameter less than 2.5 microns (PM2.5). The amount of PM produced per unit area was then calculated by multiplying the concentration once it stabilized (when it became uniformly mixed) by the volume of the enclosure and dividing by the area treated. To directly compare the PM emission characteristics of blowing, vacuuming, raking, and sweeping, the surface to be treated in the enclosure was loaded with surrogate debris. To develop the composition of this surrogate material, bulk samples were collected from areas on the University of California, Riverside (UCR) campus where leaf blowing was about to be conducted to determine the mass of soil and vegetative matter present where these cleaning activities are conducted. The test system was then used to measure emissions from leaf blowing over surfaces where leaf blowing is typically conducted and over surfaces where a surrogate mixture of soil (obtained from the San Joaquin Valley) and vegetative matter was deposited by our staff. A number of emission tests were performed using the natural/indigenous material at the CE-CERT facility in Riverside and at the UC Kearney Agricultural Center in Parlier, CA.

<u>Health Effects of Airborne Fine Particles ($PM_{2.5}$) and Ozone</u> The goal of this study (Tyner, 2007) is to determine whether exposures to airborne fine particles ($PM_{2.5}$) and ozone (O_3) induce lung inflammation that is related to exacerbations and severity of

disease in nonatopic asthmatics and, reflecting the multi-factorial web of causation of asthma, how this relation is modified by viral infection and genetic predisposition. The development of urinary PAH-quinone biomarkers to monitor individual exposure to $PM_{2.5}$ provides an enhanced method for examining associations between exposures and health outcomes. In addition, measurement of organic species (2-3 ring PAH and oxygen derivatives) and wood-smoke tracers (levoglucosan) in ambient $PM_{2.5}$ samples and subject urine samples provides information on the impacts of PM composition and reactivity on respiratory health and potential associations with exacerbation of asthma.

Ambient 24-hr PM_{2.5} filter samples will be obtained from the Clovis and Winery monitoring sites throughout the study period and analyzed by project investigators (Hasson Lab). Filter samples will be collected 1-in-3 days from October 2007 – March 2008 and 1-in-6 days from April 2008– September 2008 per US EPA PM_{2.5} monitoring schedule. Following gravimetric analysis by the Ventura County Air Pollution Control District Lab, filters will be archived for 30 days (stored under controlled temperature/humidity) after which filters will be returned to the District. Filters will then be retrieved from the District by project investigators and filters will be extracted in organic solvent and solute composition (PAH, PAH-quinone, levoglucosan) and reactivity (ROS generation) will be determined.

3.e.1 Recent Particulate Research Study References

- Fitz, D. R., Pankratz D., Chitjian, M., Bristow. J., Peterson S. (2006) Particulate Matter Emissions Factors and Emissions Inventory from Leaf Blowers in use in the San Joaquin Valley, *CE-CERT, UC Riverside*
- Lighthall, D (2007) Email communication
- Tyner, T. (2007) Email communication

Additional links to peer-reviewed publications that complement the staff reports and memos cited in this Appendix:

Basic Characterization:

http://www.arb.ca.gov/airways/CRPAQS/PM_DA/final/Chowetal2006JGRChemC ompSpatioVar.pdf

Fog:

http://www.arb.ca.gov/airways/CRPAQS/PM_DA/final/Herckesetal2005JAWMAF ogOverview.pdf

Secondary Aerosols:

http://www.arb.ca.gov/airways/CRPAQS/PM_DA/final/Lurmannetal2006JAWMAS_JVFresnoSecAerosol.pdf

Transport:

http://www.arb.ca.gov/airways/CRPAQS/PM_DA/final/MacDonaldetal2006JAWM ASJVPMTransport.pdf