Response to Reviewers for “Chemistry of new particle growth in mixed urban and biogenic emissions - Insights from CARES”

We thank the two anonymous reviewers for their comments, which have helped us to improve the paper. Our specific responses can be found below, with reviewer comments in black and our responses in blue.

Reviewer #1

This manuscript describes particle microphysical and chemical properties during new particle events (NPEs) that occurred in an urban-influenced forested site in California. The principal finding is that NPEs occurred primarily under urban transport; new particle formation and growth did not occur in the absence of an anthropogenic trigger. This finding is in contrast with the extensive work on new particle formation and growth in the forests of Finland, where no anthropogenic component is required for frequent regional NPEs. In both cases the growth is primarily due to organic composition, yet in the California case NPEs happen only when the urban plume is over the measurement site.

The manuscript is well written and of interest to ACP readers. I have some suggestions to shorten the manuscript a bit; I recommend it be accepted with minor modification.

Suggested Changes (page numbers and line numbers refer to "printer-friendly" version of manuscript):

1) The Experimental section describes the facilities at the urban (T0) site and the forested (T1) site. Nowhere is it explicitly stated that chemical composition was measured only at the T1 site, while size distributions were measured at both. This is a problem because later in the paper the chemical data are introduced without stating clearly that the measurements are from the T1 site, which caused me some confusion at first.

We already mentioned in the section 2.1 (Sampling site and instrumentation) that the AMS data was that of T1 (from page 2049 line 23 to page 2050 line 8), and that the SMPS data were from both T0 and T1 (page 2050, from line 9 to 22). However, in the revised manuscript, we have added a sentence in the Experimental section to further clarify this point.

2) P. 2051, lines 5-12, there is a discussion about thresholds for discriminating "strong" from "weak" NPE cases. There is no mention of the time required for the stated increase – is it 800 particles/cm^3/hr? Why is this inconsistent with the number increase shown in Table 1, where the "strong" and "weak" cases are shown? This needs to be clarified both here and in Table 1.

Indeed, our classification does not take into account the time needed for the increase of the particle concentration. The threshold to consider an event as NPE is an increase by 800 #/cm^3 (not 800 #/cm^3/hr), while the threshold between “weak” and “strong” NPE is an increase by 1500 #/cm^3. In this classification, we consider only particles in the range 12-20 nm (in D_m). We agree with the reviewer that the discussion is confusing, because the classification of “strong” and “weak” events presented in the text does not correspond to the data given in Table 1 (ΔN, expressed in #/cm^3/hr). Therefore, we renamed the two columns “ΔN” in Table 1 as “ΔN/Δt, and we included two new columns corresponding to $\Delta N_{12\text{-}20\,\mathrm{nm}}$, which is consistent with the discussion in the text.

3) At several locations in the paper (e.g., p. 2051, line 14), the word "mode" is used in place of "modal diameter" or "number geometric mean diameter". "Mode" means the entire aerosol population in a given size range. Please search and correct throughout the text.

We thank the reviewer for pointing this out. We now use the term “modal diameter” throughout the manuscript.

4) p. 2052, line7, it's not clear that these NPEs occur on a regional scale. They are occurring in the urban center and at a downwind site; does this constitute "regional scale" or "plume scale"?

T0 and T1 are separated by ~40 km. The fact that the SMPS data showed "banana shapes" at both locations suggests that these NPEs occurred on a regional scale.

5) p. 2052, line 16. Are the differences in the growth rates at T0 and T1 statistically significant?

We performed a Student's t-test to check the significance of the mean values of the growth rates. The difference in the mean growth rate at T0 (7.1 ± 2.7 nm h^{-1}) and T1 (6.2 ± 2.5 nm h^{-1}) is indeed not statistically significant (i.e., $p>0.05$). However, it is generally true that the growth rate at T0 is higher than at T1 for individual days. This point is now clarified in the manuscript.

6) p. 2053, line 1. Use "T0" and "T1" consistently instead of "Cool" and "Sacramento".

Done.

7) p. 2053, line 7. A nucleation mode is smaller than 10 nm; you are measuring the Aitken mode.

We corrected this point in the manuscript.

8) p. 2053, line 19. By agglomeration do you mean coagulation? Self-coagulation rates are probably very low; growth in the Aitken mode is vastly dominated by condensation in these circumstances of rapid diameter increase.

We agree that coagulation rates are likely very low during these events. We included it in the manuscript and correct the sentence.

9) Recommend removing Fig. 3, as it shows the same information as in Fig. 2. Do you really need both?

We thank the reviewer for this suggestion, but we think that size distributions shown in Fig. 3 are still useful to visualize more quantitatively the apparition of the Aitken mode at 9:00 (T0) and at 11:00 (T1), and also the difference of particle concentrations at the two sites. Therefore, we prefer to keep this figure in the manuscript.

10) p. 2054, it would be very useful to show the condensation sink term on one of the diurnally averaged plots, like Fig. 4 or Fig. 5. The sink term may play as big a role in determining NPEs as does the source term.

We now include the diurnal patterns of condensational sink in Fig. S4.

11) p. 2059, lines 5-8. The compounds you list are not semivolatile and will not partition to the particle phase. They are rather markers of oxidation that are probably correlated with condensable compounds.

We thank the reviewer for pointing this out. The sentence has been modified accordingly.

12) p. 2059 lines 14-18. You haven't shown that the differences in these compounds between NPE days and non-NPE days are significant. The standard deviations certainly overlap. Use "significantly" only when you've done the statistical testing to verify.

Standard deviation indicates the spread of the data points from the mean. Since ambient concentrations vary from time to time, the fact that the (mean ± standard deviation) values for NPE and non-NPE days overlap doesn't necessarily indicate the differences are statistically insignificant. We thus performed Student's t-test and determined the significance of the mean values. Among the anthropogenic species mentioned on p. 2059 lines 14-18, the difference between NPE and non-NPE is significant (i.e., $p<0.05$) for BC, CO, HOA and toluene. NO_x is the only species for which the mean values are not statistically different. We have revised the texts accordingly.

13) p. 2059 line 26 through p. 2060 line 13. This is a long discussion for a very minor issue (RH); can it be shortened to, "there was no evident relationship between measured RH and particle growth rates or the occurrence of NPEs"?

In response to the reviewer's comments, we have shortened this paragraph in the revised manuscript. However, we think that this discussion is important, because previous studies already focused on the influence of RH on nucleation events, and for the moment, the role played by RH on these events is not clear yet.

14) p. 2061 line 24, again, are these growth rate differences really significant?

Based on Student's t-test, the differences in the mean values of the growth rates at T0 and T1 are not statistically significant. We have revised the text accordingly.

15) Figure S5. It would be nice to add a trace of CO to one of these plots so that we can see the urban influence in a non-aerosol tracer. Ozone is good but is secondary and regional, while NO_x gets converted to NO_y and is not a great tracer.

The time series of CO is now included in this graph.

Technical Edits:

1) p. 2058, "NPE events" should just be "NPEs".

We replaced "NPE events" with "NPEs" throughout the manuscript.

2) References, several place names (e.g., Texas, New England) need to be capitalized. Please thoroughly check the references for typos; I have not.

We checked all the references, and corrected the place names.

3) Table 1, column labeled "deltaN" is inconsistent with text describing the "strong"/"weak" classification. Is this a different particle size range than described in the text? Please state what this variable is.

In Table 1, ΔN corresponds to the difference of the particle concentration between the end and the beginning of the growth, normalized to the time, and takes into account all the particles in the range 12-737 nm (for both T0 and T1). We have revised Table 1: 1) rename these two columns to "ΔN/Δt" and 2) include two new columns corresponding to "$\Delta N_{12\text{-}20\ \mathrm{nm}}$". Our classification of "strong" and "weak" events is based on $\Delta N_{12\text{-}20\ \mathrm{nm}}$, thus the discussion in the text and the data in Table 1 are now consistent.

4) Fig. 6 caption, state clearly what the lines+symbols are.

We have clarified in the figure caption that the stacked curves correspond to the % contribution (left axes), while the lines+symbols correspond to the total masses (right axes).

5) Fig. 9 caption. You don't need to define NPE here again. Missing the right-hand column of graphs (d-f)!

We apologize for the wrong figure caption, which refers to a previous version of this graph. We corrected the figure caption.

6) Change "NPG" to "NPE" in this figure.

Done.

7) Supplemental material p. 2, line 22. Define "PToF".

Done.

8) Supplemental material p. 2, line 23. Change "data of ammonium was" to "data of ammonium were".

Done.

9) Figure S1. I have no idea what the various lines, symbols, and bars on this figure mean, or what it's supposed to show. Is this only for those who process AMS data?

We now include a legend on this panel to explain what the various lines mean.

Reviewer #2

This paper discusses aerosol observations during June 2010 at two Californian sites: an urban site in the Sacramento, CA and a rural site about 40 km northeast of Sacramento. The observations focus on the growth of newly formed particles observed on several days at both sites, the connections between the sites and the chemistry of the particles. The new particle growth events are found to be associated with the transport of the aerosol from the urban area towards the rural site. The authors suggest that most of the growth was due to anthropogenic SOA, and there is a link between the growth event and the presence of amines in the particles. The consistency between the AMS mass concentrations and MPSS volume concentrations down to near 30 nm VAD is remarkable (Fig. 8). The presentation is well done. I have a few minor comments for the authors.

Specific comments:

1) I believe that SMPS is a trade mark of TSI, and thus the authors may wish to avoid its generic use here. Why not use MPSS?

We followed the suggestion of the reviewer and used the term “MPSS” throughout the manuscript.

2) Was the AMS lens a standard lens or is it a new design? It is unclear from Setyan et al (2012). What is the reference for its characterization?

The aerodynamic lens of our AMS is a standard lens, described in Zhang et al. (2004). It is now included in the revised paper.

3) Page 2015 – Are all the distributions for which you are estimating the mode diameters log normal?

Yes, the mode diameters are always determined with log normal size distributions. We clarified this point in the manuscript.

4) Page 2052, line 25 - Page 2053, line 5 – How do your growth rates depend on your somewhat arbitrary definition of “when the growth significantly slows down”? Increased biogenic precursor concentrations could result in larger growth rates. The growth rate in a biogenic environment also depends on the volatility of the condensing material, a point discussed by Riipinen et al (ACP, 2011), Pierce et al (ACP, 2011), Pierce et al (ACP, 2012) and most recently in a Nature publication. These additional points should be mentioned here.

Typically, the growth rate is quite linear during the first 2-3 hours, then it slows down by a factor of ~50% during the following 2-3 hours, and decreases to 0 nm/hr ~8 hours after the start of the observation. One reason for the decrease of the growth rate after a few hours may be due to the fact that when particles grow to a certain diameter, the condensation of additional species onto the surface of these particles will result in a very small increase of their sizes. The reviewer is right to point out that the concentration and volatility of the condensing material may also influence the growth rate. We included this discussion in the manuscript and cited the references.

5) Page 2053-2054 and Figures 2-4 – It is worth pointing out here that the delay between the two sites and the absence of particles smaller than 20 nm compared with T0, suggests that the particle nucleation occurred much near and upwind of T0 and not close to T1. In other words, the banana observed at T1 was likely independent of the emissions in the T1 area and mostly dependent on the emissions near T0 and upwind of T0. That is of course consistent with your general conclusion that the growth was dominated by anthropogenic precursors.

We agree with the reviewer, and this point is mentioned in the manuscript.

6) What about the seemingly independent mode bounded by about 11am and 5 pm and 60-100 nm in Figure 2? Was that common during the NPE days, and how did that mode influence your estimate of the composition of the 40-120 nm particles?

Particles in the range 60-100 nm (in D_m) between 11:00 and 17:00 seem to correspond to particles formed during the previous NPE. This mode was observed very often during nighttime throughout this study, as shown in the diurnal pattern of Fig. 4c. It is a bit difficult to assess the influence of this mode in the particle chemistry in the range 40-120 nm (in D_{va}), because we would need to study the particle chemistry in two different size ranges, e.g. 40-80 nm and 80-120 nm (in D_{va}). Since the PToF data of the AMS species tend to be noisy due to low duty cycle under PToF mode, it will be difficult to obtain useable results without averaging a sufficient number of size bins.

7) Page 2057, lines 12-13 – How were the cases of "dominate biogenic influence" derived?

The periods of biogenic influences were determined with the North American Mesoscale (NAM) model, and given in Fast et al. (2012). The model determined 3 periods (June 10-13, 16-17 and 20-21) during which the wind direction shifted from southwest to north/northwest, due to a trough passing over California. The northwest region of T1 is heavily forested and has a lot of agricultural areas, but without any significant urban areas. The biogenic SOA was more concentrated than the anthropogenic SOA during these 3 periods (Setyan et al., 2012).

8) Page 2057, lines 21-23 and Fig 7c – Despite the highest temperatures from 10 am to 4 pm, a 10% increase in biogenic SOA across that time is sensible compared with previous observations of BSOA (e.g. Slowik et al., 2010; Pierce et al., 2012). The spatial scales of anthropogenic and biogenic emissions are so much different, and your results seem to be an excellent demonstration that the anthropogenic components dominate on smaller scales.

We agree with the reviewer.

9) Section 3.3 and Figure 10 – Was SO_2 measured? Figure 10 appears to need an "(a)" caption.

SO_2 was not measured at T1. The caption of the (a) panel of Fig. 10 is already there (last sentence of the figure caption).

10) Page 2060, lines 6-9 – I am confused by your apparent conclusion in this sentence. Are not sunny days those that would have the solar radiation peak about noon?

During sunny days, the solar radiation peaks at noon and reaches 1000-1200 W/m^2. When the weather is cloudy, the solar radiation usually also peaks at noon, but reaches much lower values.

We modified the sentence in line 8-10, which now reads: "However, in our case, this does not seem to explain the different behavior of RH between NPE and non-NPE days, since the weather was sunny during the entire field campaign."

11) Page 2061, lines 5-15 and page 2062, lines 14-16 – If your results show that biogenic SOA was a small contributor to the growth, what is the basis for saying that growth was promoted by the interaction of urban and biogenic emissions?

T1 is located in a forested region where biogenic influences are always significantly present. As shown in Fig. 10, the average isoprene concentrations were almost identical during NPE and non-NPE days but the first generational products of isoprene oxidation (MACR+MVK) were ~ 20% higher during NPE days. Biogenic SOA, although a smaller contributor to the growth compared to urban transport SOA, its concentration was ~ 50% higher during NPE days than during non-NPE days. In addition, species representative of anthropogenic emissions, including BC, CO, benzene, and toluene, all increased by 30 – 60% during NPE days, indicating the importance of the anthropogenic influence on the occurrence of NPE events. Further, the average concentration of urban transport SOA more than doubled during NPE days. These results together indicate that the growth was promoted by the interaction of urban and biogenic emissions.

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Chemistry of new particle growth in mixed urban and biogenic emissions - Insights from CARES

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Abstract

Regional new particle formation and growth events (NPE) were observed on most days over the Sacramento and western Sierra Foothills area of California in June 2010 during the Carbonaceous Aerosols and Radiative Effect Study (CARES). Simultaneous particle measurements at both the T0 (Sacramento, urban site) and the T1 (Cool, rural site located ~40 km northeast of Sacramento) sites of CARES indicate that the NPE usually occurred in the morning with the appearance of an ultrafine mode ~~centered~~ at ~15 nm (in mobility diameter, D_m, measured by a ~~scanning~~ mobility particle ~~sizer~~size spectrometer operating in the range 10-858 nm) followed by the growth of this ~~mode~~modal diameter to ~50 nm in the afternoon. These events were generally associated with southwesterly winds bringing urban plumes from Sacramento to the T1 site. The growth rate was on average higher at T0 (7.1±2.7 nm/hr) than at T1 (6.2±2.5 nm/hr), likely due to stronger anthropogenic influences at T0. Using a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS), we investigated the evolution of the size-resolved chemical composition of new particles at T1. Our results indicate that the growth of new particles was driven primarily by the condensation of oxygenated organic species and, to a lesser extent, ammonium sulfate. New particles appear to be fully neutralized during growth, consistent with high NH_3 concentration in the region. Nitrogen-containing organic ions (i.e., CHN^+, CH_4N^+, $C_2H_3N^+$, and $C_2H_4N^+$) that are indicative of the presence of alkyl-amine species in submicrometer particles enhanced significantly during the NPE days, suggesting that amines might have played a role in these events. Our results also indicate that the bulk composition of the ultrafine mode organics during NPE was very similar to that of anthropogenically-influenced secondary organic aerosol (SOA) observed in transported urban plumes. In addition, the concentrations of species representative of urban emissions (e.g., black carbon, CO, NO_x, and toluene) were significantly higher whereas the photo-oxidation products of biogenic VOC and the biogenically-influenced SOA also increased moderately during the NPE days compared to the non-event days. These results indicate that the frequently occurring NPE over the Sacramento and Sierra Nevada regions were mainly driven by urban plumes from Sacramento and the San Francisco Bay Area and that the interaction of regional biogenic emissions with the urban plumes has enhanced the new particle growth. This finding has important implication for quantifying the climate impacts of NPE on global scale.

1 Introduction

New particle formation and growth processes are an important source of ultrafine particles in both clean and polluted environments. A large number of studies reported the observations of intensive new particle events at various locations, including urban areas (e.g., Brock et al., 2003; Dunn et al., 2004; Stanier et al., 2004; Zhang et al., ~~2004~~2004a; Wu et al., 2007; Ahlm et al., 2012), remote sites (e.g., Weber et al., 1999; Creamean et al., 2011; Vakkari et al., 2011; Pikridas et al., 2012), forested locations (e.g., Allan et al., 2006; Pierce et al., 2012; Han et al., 2013), coastal sites (e.g., O'Dowd et al., 2002; Wen et al., 2006; Liu et al., 2008; Modini et al., 2009), and polar regions (e.g., Komppula et al., 2003; Koponen et al., 2003; Asmi et al., 2010). These events significantly affect the number concentrations and size distributions of particles in the atmosphere with important implications on human health and climate (Spracklen et al., 2006; Bzdek and Johnston, 2010; Kerminen et al., 2012). However, despite frequent observations, the chemical processes underlying the formation and growth of new particles remain poorly understood.

New particle events occur in two steps, i.e., the formation of nuclei, followed by the growth of the stable clusters to larger sizes by condensation of low-volatility compounds and coagulation. For ambient measurements, the evolution of the number-based particle size distribution is a main criterion for identifying the onset of new particle events. ~~Scanning mobility particle sizer (SMPS), also called mobility~~Mobility particle size spectrometer (MPSS), also called scanning mobility particle sizer (SMPS), is the most widely used instrument to determine the particle number concentration and size distribution during these events. The evolution of the chemical composition of ultrafine particles during new particle formation and growth is another piece of critical information needed for understanding this process. For that purpose, aerosol mass spectrometer (AMS) (e.g., Zhang et al., ~~2004~~2004a; Allan et al., 2006; Ziemba et al., 2010; Creamean et al., 2011; Ahlm et al., 2012), chemical ionization mass spectrometer (CIMS) (e.g., Dunn et al., 2004; Smith et al., 2005; Smith et al., 2008; Smith et al., 2010; Jokinen et al., 2012), Nano aerosol mass spectrometer (NAMS) (e.g., Bzdek et al., 2011; Bzdek et al., 2012), and atmospheric pressure ionization time-of-flight (APi-TOF) mass spectrometer (Lehtipalo et al., 2011; Kulmala et al., 2013) have been successfully deployed in the field to study the chemical processes underlying atmospheric new particle events.

An important finding from previous studies is that organics and sulfates are usually involved in the growth of new particles up to sizes where they can act as cloud condensation nuclei (CCN). The contribution of these two species to particle growth depends on the concentrations of the precursors and meteorological conditions. For example, at urban or industrial locations where the SO_2 mixing ratio is high, sulfate is an important contributor to the growth of new particles (Brock et al., 2003; Zhang et al., ~~2004~~2004a; Yue et al., 2010; Bzdek et al., 2012). At rural and remote locations, however, the growth of new particles was found to be almost exclusively driven by organics (Smith et al., 2008; Laaksonen et al., 2008; Ziemba et al., 2010; Pierce et al., 2012; Ahlm et al., 2012). In addition, it was found that in Pittsburgh, USA, despite high ambient SO_2 concentrations, H_2SO_4 contributes mainly to the early stage of the new particle growth, while the growth up to CCN sizes is mainly driven by secondary organic aerosols (SOA), especially during late morning and afternoon when photochemistry is more intense (Zhang et al., ~~2004~~2004a; Zhang et al., 2005).

SOA is a major component of fine particles globally (Zhang et al., 2007; Jimenez et al., 2009). Understanding its roles in new particle formation and growth is important for addressing aerosols' effects on climate and human health. Recent studies found significantly enhanced SOA formation rates in mixed biogenic and anthropogenic emissions (de Gouw et al., 2005; Volkamer et al., 2006; Kleinman et al., 2008; Setyan et al., 2012; Shilling et al., 2013). However, there is little known about the influence of the interactions of organic species from biogenic and anthropogenic sources on new particle growth. The Sacramento Valley in California is a place of choice to study this process. The Sacramento metropolitan area lies in the Central Valley to the north of the San Joaquin River Delta and to the southwest of the forested Sierra Nevada Mountains. The wind in this region is characterized by a very regular pattern, especially in summer (Fast et al., 2012). Indeed, during the day, a southwesterly wind usually brings air masses from the San Francisco Bay to the Sacramento metropolitan area and pushes northeast to the Sierra Nevada Mountains (Dillon et al., 2002), promoting the transport of urban plumes from Sacramento to forested regions where biogenic emissions are intense.

The U.S. Department of Energy (DOE) sponsored Carbonaceous Aerosols and Radiative Effects Study (CARES) that took place in the Sacramento Valley in June 2010 was designed to take advantage of this regular wind pattern to better understand the life-cycle processes and radiative properties of carbonaceous aerosols in a region influenced by both anthropogenic and biogenic emissions (Zaveri et al., 2012). Within the framework of CARES, a wide range of instruments were deployed between June 2 and 28, 2010 at two ground sites located in Sacramento (T0, urban site) and Cool, CA at the foothills of the Sierra Nevada Mountains

(T1, rural site), respectively, to measure size-resolved chemical compositions, number size distributions, and optical and hygroscopic properties of aerosols, as well as trace gases and meteorological data (Zaveri et al., 2012). One of the major observations during CARES was that particles were dominated by organics in this region, and that the formation of SOA was enhanced when anthropogenic emissions from the Sacramento metropolitan area and the Bay Area were transported to the foothills and mixed with biogenic emissions (Setyan et al., 2012; Shilling et al., 2013).

During CARES, new particle growth events were observed almost daily at both the T0 and T1 sites. Similarly, previous studies conducted at the University of California Blodgett Forest Research Station, approximately 75 km to the northeast of Sacramento and 35 km to the northeast of the T1 site, also reported the frequent occurrence of NPE (Lunden et al., 2006; Creamean et al., 2011). In their study conducted from May to September 2002, Lunden et al. (2006) found that the oxidation products of reactive biogenic compounds accounted for a significant portion of the particle growth. The study of Creamean et al. (2011), which took place in early spring of 2009, found that sulfates and amines participated in the growth of new particles and that long-range transport of SO_2 from Asia seemed to contribute to faster growth. These findings indicate that new particle formation and growth are important processes in Northern California and are affected by regional anthropogenic and biogenic emissions as well as by pollutants transported from Asia. Understanding ~~how~~to what extent these emissions may govern the NPE's requires measurements of size-resolved chemical compositions of the new particles. The main aim of the present paper is to examine the evolution characteristics of new particles at the T0 and T1 sites during CARES, with a focus on the evolution of size-resolved particle chemical composition based on HR-ToF-AMS measurements at T1.

2 Experimental

2.1 Sampling site and instrumentation

The T0 sampling site was located on the campus of the American River College in Sacramento (38° 39′ 01” N, 121° 20′ 49” W, 30 m above sea level) and the T1 site was located on the campus of the Northside School at Cool (38° 52′ 16” N, 121° 01′ 22” W, 450 m above sea level). Sacramento is the capital of California, with 480,000 inhabitants in the city and 2.5 million people living in the metropolitan area. Cool is a small town (2500 inhabitants) surrounded by very large forested areas, and located ~40 km northeast of Sacramento at the Sierra Nevada foothills.

In this paper, we report results of particle chemical compositions at T1, and particle number size distributions at both T0 and T1. Size-resolved chemical composition of non-refractory submicron aerosols (NR-PM$_1$) were measured at T1 using an Aerodyne HR-AMS (DeCarlo et al., 2006; Canagaratna et al., 2007). A detailed discussion on its operation ~~at T1~~ during the present study was presented in Setyan et al. (2012)~~. Briefly, the HR-AMS was equipped with an aerodynamic lens~~. Briefly, the HR-AMS was equipped with a standard aerodynamic lens, described in Zhang et al. (2004b), and allowing the transmission of particles in the range ~30-1500 nm (in vacuum aerodynamic diameter, D_{va}). The instrument was operated alternatively in V- and W-mode every 2.5 min. In V-mode, data was recorded in mass spectrum (MS) mode and particle time-of-flight (PToF) mode. The MS mode was used to obtain average mass spectra and determine the concentration of the species in submicrometer particles without size information. In the PToF mode, average mass spectra were acquired for 92 size bins covering 30-1500 nm (D_{va}), allowing the determination of the size-resolved chemical composition. W-mode data was recorded exclusively in MS mode.

The particle number size distribution was measured both at T0 and T1 with a ~~SMPS~~MPSS (also called ~~MPSS, type TROPOS~~SMPS) as described in Wiedensohler et al. (2012). The instrument used at T1 consists of a Hauke-type differential mobility analyzer (DMA) and a condensation particle counter (CPC; TSI Inc., Shoreview, MN; model 3772), and used ^{210}Po as radioactive source for the neutralizer (Setyan et al., 2012). The ~~SMPS~~MPSS was set to measure particles in the range 10-858 nm (in mobility diameter, D_m), divided into 70 logarithmically distributed size bins. ~~SMPS~~MPSS data has been corrected to take into account the DMA-CPC lag time, bipolar charge distribution, CPC efficiency, and diffusion loss. The SMPS deployed at T0 was a commercial instrument (TSI Inc.; model 3936), and was constituted of a ^{85}Kr neutralizer, a DMA (TSI Inc.; model 3080 with the long column) and a CPC (TSI Inc.; model 3775). The instrument measured particles in the size range of 12-737 nm (in D_m) divided into 115 size bins. Diffusion loss correction was applied after the data inversion. All dates and times reported in this paper are in Pacific Daylight Time (PDT = UTC – 7 hr), which was the local time during this study.

2.2 Data analysis

Particle number concentration and size distribution have been used to identify new particle events in the atmosphere. However, given that the new particles formed by nucleation have generally a diameter in the size range 1-3 nm, smaller than the smallest size measured by our ~~SMPS's~~MPSS's, we were not able to observe the new particle formation themselves during the present study, but only the growth of the newly formed particles that are larger than 10 nm. For this reason, we will not use the terms "nucleation" or "new particle formation" in the forthcoming discussion, but rather "new particle growth". Each day for which complete ~~SMPS~~MPSS data was available was classified as new particle event (NPE) day if the particle number concentration in the size range 12-20 nm increased by more than 800 particles/cm^3, and if this increase was accompanied by the increase of the ~~mode~~modal diameter during the following hours. These two conditions allowed us to distinguish NPE from primary emissions from vehicles, which also produce small particles but are usually observed as occasional spikes in the time series of the particle number concentration in the range 12-20 nm. In addition, each growth event was considered as "strong" if the increase of the particle number concentration in the range 12-20 nm was higher than 1500 particles/cm^3, and "weak" if the increase was lower than this threshold. A summary of the new particle growth events observed during this study is provided in Table 1.

The ~~mode~~modal diameter(s) of each particle number size distribution recorded during this study have been determined with a multiple peak fitting tool available in Igor Pro 6.2.2.2 (WaveMetrics Inc., Lake Oswego, OR). All the size distributions were log normal. The growth rate (GR), which corresponds to the increase of the ~~mode~~modal diameter of newly formed particles per time unit (nm/hr), has been calculated for each individual growth event using Equation 1:

$$GR = \frac{\Delta Dm}{\Delta t} \qquad (1)$$

in which ΔD_m is the difference of the ~~mode~~modal diameter (nm) between the beginning of the growth and the period when the growth significantly slows down, and Δt is the duration of the growth (hr).

3 Results and discussions

3.1 Evolution of particle number size distributions during regional new particle events

The ~~SMPSs~~SMPS and MPSS were fully operational during 26 days at T0, and 22 days at T1, from June 2 – 29, 2010. The time series of the particle number size distributions show that new particle events frequently occurred at both sites (Fig. 1), indicating that these events occurred on a regional scale. A total of 19 NPE were identified at T1 (86% of the time; Table 1), eight of which were considered as "strong" and eleven as "weak". Most of the events (14 in total) occurred during periods of southwesterly wind that transported urban plumes to the T1 site (i.e., T0 → T1), except for 5 events which occurred during northwesterly wind periods

(Table 1). In addition, all 8 strong NPE occurred during the T0 → T1 periods (Table 1). At T0, 22 new particle events were identified, 18 of which were considered as "strong" and only four events were "weak".

Fig. 2 compares the average daily evolution patterns of particle number concentrations at the T0 and T1 sites during NPE days. Generally, the increase of the particle number concentration during these events was significantly higher at T0 than at T1 (average 9.~~6E3~~$6\cdot10^3$ vs. 3.~~8E3~~$8\cdot10^3$ #/cm^3/hr, $p<0.05$ with Student's t-test; Table 1~~) and the~~). The average (± 1σ) growth rate of new particles was ~~slightly~~also higher at T0 (7.1 ± 2.7 nm/hr vs. 6.2 ± 2.5 nm/hr at T1~~). The~~), but the difference was not statistically significant (i.e., $p>0.05$ with Student's t-test). The growth rates given in Table 1 correspond to the first hours of the observation, when the increase of the modal diameter is linear. Indeed, the growth rate is usually quite linear during the first 2-3 hours and slows down afterwards (Fig. 5a and 5c). One reason for the decrease of the growth rate after a few hours may be due to the fact that when particles grow to a certain diameter, the condensation of additional species onto the surface of these particles will result in a very small increase of their sizes. The occurrence of relatively stronger ~~and faster~~ NPE at T0 is likely due to the proximity of emission sources of precursor species and a higher anthropogenic influence. Indeed, the frequency as well as the growth rates observed during the present study were much higher than those reported by Lunden et al. (2006) at ~35 km northeast of T1 (frequency = 30% of the time, average growth rate = 3.8 ± 1.9 nm/hr), where the lower frequency and growth rates might be related to the fact that their site was located deeper into the forest and subjected to relatively lesser anthropogenic influences from urban areas to the southwest (e.g., Sacramento and the San Francisco Bay Area). The growth rates measured during the present study are also much higher than those observed at Hyytiälä, Finland, where NPE have been extensively observed and described over the past 15 years. Riipinen et al. (2011) report a median growth rate of 2.3 nm/hr during the years 2003-2007, much lower than at ~~Cool~~T1 (6.2 nm/hr) and ~~Sacramento~~T0 (7.1 nm/hr). NPE at Hyytiälä are mainly driven by the photooxidation of biogenic precursors~~.~~, and thus growth rates measured in this kind of environment depend on the concentration and volatility of the condensing material (Pierce et al., 2011; Riipinen et al., 2011; Pierce et al., 2012; Riipinen et al., 2012). The Sacramento and Sierra Foothill region, however, is influenced by both urban and biogenic emission sources. Thus, the comparison between the growth rates at these different sites suggests that the degree of anthropogenic influence may be an important factor driving the growth rate.

During the present study, all growth events began in the morning, with the appearance of ~~a nucleation~~an Aitken mode observed with the ~~SMPS~~MPSS between 9:00 and 12:00 (PDT). Particle growth lasted several hours, with size modes reaching their maximum in the afternoon, typically after 15:00. The ~~modes~~modal diameters at the end of the growth in general peaked between 40-50 nm, but for several cases, the ~~mode~~modal diameter did not reach 35 nm, especially for the weakest events or when a change in the wind direction was observed during the day (Fig. 1).

An important observation of the present study is that NPE began at T1 a few hours later than at T0, especially during days characterized with daytime T0 → T1 transport. A typical example of this phenomenon occurred on June 26 (Fig. ~~2~~3 and ~~3~~4). According to Fast et al. (2012) , a T0 to T1 transport occurred that day. Particles smaller than 20 nm (in D_m) began to increase slightly before 9:00 at T0 (Fig. ~~2a~~3a), and ~~a nucleation~~an Aitken mode appeared at the same time (Fig. ~~3~~4). Then, during the following hours, the ~~mode~~modal diameter increased slowly up to ~50 nm (in D_m), likely due to ~~agglomeration and~~ condensation of low-volatility compounds onto the surface of these new particles. The increase of the modal diameter could also be due to coagulation, but this process is expected to be very slow for particles in the Aitken mode. Thus, as shown in Fig. ~~2a~~3a, the evolution of the particle number size distribution shows a "banana shape", which is a typical observation ~~with the SMPS~~ for the growth of new particles. At T1, the same phenomenon occurred at ~11:00, i.e., 2 hours after T0 (Fig. ~~2b~~3b). This time delay is consistent with the wind data recorded at T1 which indicate the sampling of air masses transported from the T0 direction. The much lower concentrations of particles smaller than 20 nm between 9:00 and 11:00 at T1 (Fig. 3b), compared with T0 (Fig. 3a), suggests that new particle formation occurred much near and upwind of T0 and not close to T1. In other words, the banana-shaped evolution pattern observed at T1 was likely independent of the emissions in the T1 area and mostly dependent on the emissions near T0 and upwind of T0. Further evidence for this pseudo Lagrangian sampling is the observation of a sudden change in wind direction at ~14:30 at T0 that brought in a very clean air mass associated with a sharp decrease of particle number concentration that lasted for ~3.5 hours (Fig. ~~2a~~3a). Particle concentration at T0 increased again at ~18:00 after a shift of the wind direction back to southwesterly. A mirrored decrease of particle concentration, although less dramatically, was observed at 16:30 at T1, ~2 hours after the clean air mass event at T0 (Fig. ~~2b~~3b). The increase of particle number concentration occurred at T1 around 21:00, ~3 hours after the increase occurred at T1, consistent with gradually decreasing wind speed from 16:30

to 21:00. The wind direction at T1 remained southwesterly during the entire afternoon (Fig. ~~2b~~3b).

This time delay between T0 and T1 was also observed during the other events, and this is confirmed by the diurnal evolution profiles of ~~the~~ particle number concentrations (Fig. 2) and size distributions at both sites (Fig. ~~4a~~5a and ~~4c~~5c). These observations indicate that new particle growth generally occurred during T0 → T1 transport promoted by the daytime southwesterly wind and that the new particle growth events were generally more intense at T0 compared to at T1. Wind rose plot during NPE (Fig. ~~5g~~6g) confirms that these events usually occurred when the wind was coming from the southwest, which corresponds to the location of the Sacramento metropolitan area. On the other hand, when NPE was not observed, the wind was coming mainly from the northwest and the west (Fig. ~~5h~~6h), bringing air masses dominated by biogenic emissions (Setyan et al., 2012), thus reducing anthropogenic influences at T1.

It is interesting to notice that the evolution of the particle number size distributions and concentrations during the evening and the night is not similar at T0 and T1. At T0, particle number concentration remains almost constant between 23:00 and 8:00, while the mode is centered at ~35-40 nm (in D_m) during this period (Fig. ~~4a~~5a). On the contrary, particle number concentration decreases gradually at T1 during night, while the ~~mode~~modal diameter increases from 35 nm (at 21:00) up to 90 nm (at 14:00 the following day; Fig. ~~4c~~5c). This may be due to the fact that the T0 site was more influenced by nanoparticles from vehicular emissions than the T1 site, due to the proximity of traffic, anthropogenic emissions, and transport from the Bay Area. On the other hand, the T1 site was more influenced by downslope winds during the night, when a change in the wind direction brought down more aged aerosols from the Sierra Nevada to the foothills (Setyan et al., 2012).

3.2 Evolution of particle chemistry during new particle growth

The evolution of particle chemistry during NPE at T1 was studied in detail with a HR-ToF-AMS. As summarized in Table 1, the increase of particle number concentration during the new particle growth events was accompanied by an increase of organics and sulfate in ultrafine particles (40-120 nm in D_{va}). The average (± 1σ) increase of organics in that size range was 0.71 (± 0.29) $\mu g/m^3$ while that of sulfate was 0.10 (± 0.11) $\mu g/m^3$.

Fig. ~~5~~6 shows the diurnal size distributions of organic matter, sulfate, and particle volume concentrations, along with the wind rose plots during NPE days and non-event days. The growth of new particles was mainly contributed by sulfate and organics (Fig. ~~5a~~6a and ~~5c~~6c), but the increase of particle mass observed by the AMS occurred after 11:00, later than the increase of number concentration according to the SPMS. This is because the smallest size measured by our ~~SMPS~~MPSS is 10 nm (in D_m), while the transmission through the AMS is significant only for particles larger than 30 nm (in D_{va}) (Jayne et al., 2000). Given that particle density at T1 was on average 1.4 during this study (Setyan et al., 2012), and assuming that they are spherical, the smallest particles measured by the AMS correspond to ~21 nm in D_m. Thus, the ~~SMPS~~MPSS was the first instrument to detect the growth of new particles, while the HR-ToF-AMS observed the growth 2 or 3 hours later, depending on the growth rate. A similar observation was reported during NPE in Pittsburgh (Zhang et al., ~~2004~~2004a). It is interesting to notice that organics, sulfate, and particle volume exhibit qualitatively the same diurnal size distributions (Fig. ~~5~~6). Indeed, they have a constant ~~mode~~modal diameter in larger particles during the entire day, and they increase in ultrafine particles in the afternoon during the growth events.

The diurnal patterns of organics and sulfate in three different size ranges (40-120, 120-200, and 200-800 nm in D_{va}) show that their afternoon increase occurred mainly in ultrafine particles (40-120 nm) while the increases in the rest of the sizes were moderate during NPE days (Fig. ~~6a~~7a and ~~6c~~7c). In comparison, the diurnal profiles of both species were relatively flat and their concentrations much lower during the non-event days (Fig. ~~6b~~7b and ~~6d~~7d). Although both organics and sulfate in ultrafine particles increased in the afternoon, the increase of the organic mass in the 40-120 nm particles was on average 7 times higher than that of sulfate (see above and Fig. ~~7d~~8d and ~~7e~~8e). Clearly, the growth of new particles was mainly driven by organics. This is in agreement with previous studies, which also emphasized the key-role of organics in the growth of new particles up to CCN sizes (~~Smith~~Laaksonen et al., 2008; ~~Laaksonen~~Smith et al., 2008; Ziemba et al., 2010; Zhang et al., 2011; ~~Pierce~~Ahlm et al., 2012; ~~Ahlm~~Pierce et al., 2012; Riipinen et al., 2012).

Another important observation is the substantial increase of the signals of four nitrogen-containing ions (i.e., CHN^+, CH_4N^+, $C_2H_3N^+$, and $C_2H_4N^+$) in submicron particles during the new particle growth periods (Fig. ~~7f~~8f). On average, the concentration of these ions during NPE days was 2.4 times the concentration observed during non-NPE days (Fig. ~~10). Since these $C_xH_yN^+$ ions are generally related to alkyl-amine species (Ge et al., in preparation),~~11). Since these $C_xH_yN^+$ ions are generally related to alkyl-amine species (Ge et al., 2014), this class of compounds was likely involved in the growth of new particles. This is consistent with previous findings in the atmosphere (e.g., Makela et al., 2001; Smith et al., 2008; Smith et al., 2010; Bzdek et al., 2011; Creamean et al., 2011; Laitinen et al., 2011). Recent studies have found that sulfuric acid–amine clusters are highly stable and that even trace amount of amines (e.g., a few ppt) can enhance particle formation rates by orders of magnitude compared with ammonia (~~Almeida~~Zollner et al., ~~2013~~2012;~~Zollner~~ Almeida et al., ~~2012~~2013). The importance of gas-phase amines in the generation of organic salts involved in the formation of new particles was also confirmed by thermodynamic modeling study (Barsanti et al., 2009). Based on the mass spectrometry fragmentation patterns of amine standards analyzed in our lab (Ge et al., in preparation), the average concentration of aminium ($R_1R_2R_3N^+$, where R_1, R_2, R_3 are either H or an alkyl group) is estimated to be approximately 1/10th that of ammonium at T1 during this study (Fig. ~~7f~~8f). Although we are unable to directly assess the importance of amines in new particle formation based on this study, our results suggest that amines likely played an important role in the formation of new particles in the Sacramento and Sierra foothills region.

Due to the high contribution of organics to submicron aerosol mass in the region, positive matrix factorization (PMF) analysis was performed on the high resolution mass spectra of the AMS to investigate the sources and processes of organic aerosols (Setyan et al., 2012). Briefly, three distinct factors were determined, including a biogenically-influenced SOA associated with the regional biogenic emissions (O/C ratio = 0.54, 40% of total organic mass), an anthropogenically-influenced SOA associated with transported urban plumes (O/C ratio = 0.42, 51%), and a hydrocarbon-like organic aerosol (HOA) mainly associated with local primary emissions (O/C ratio = 0.08, 9%). Details on the determination and validation of these three OA types are given in Setyan et al. (2012). It is important to clarify here that the biogenic SOA and urban transport SOA identified at T1 do not correspond to SOAs formed from 100% anthropogenic or biogenic precursors. In fact, the so-called biogenic SOA was found in air masses with dominant biogenic influence and little anthropogenic influence, while the urban transport SOA was found in air masses characterized as urban plumes mixed with the continuously present biogenic emissions in the region. These observations are consistent with radiocarbon analysis of fine particulate matter, which has shown that modern carbon worldwide often contributes > 70% of the total carbon, particularly downwind of urban areas (Glasius et al., 2011;Schichtel et al., 2008 and references therein).

As shown in Fig. ~~7~~8, during NPE days, the mass concentration of urban transport SOA increased by more than a factor of 2 (from 0.75 to 1.7 µg/m^3) between 10:00 and 16:00 (Fig. ~~7b~~8b), whereas that of biogenic SOA increased only slightly by ~ 10% during that period (from 0.84 – 0.93 µg/m^3, Fig. ~~7e~~8c). This result underlines the key-role played by the urban plumes from Sacramento in the ~~NPE events~~NPEs at Sierra foothills.

Fig. ~~8~~9 shows the evolutions of the mass-weighted size distributions of Org, SO_4^{2-}, organic tracer ions, and particle number distributions during daytime. The average size distributions of Org and SO_4^{2-} during NPE days show significant increase of concentrations in the small mode (Fig. ~~8e~~9e and ~~8g~~9g). On the other hand, the increases of the concentrations of Org and SO_4^{2-}in ultrafine particles were all negligible during non-event days (Fig. ~~8f~~9f and ~~8h~~9h).

Another important parameter to determine was the neutralization of sulfate in the ultrafine mode during NPE. We already know from the mass spectral mode of the AMS that sulfate was fully neutralized in the bulk during the entire study (Setyan et al., 2012). Many previous studies mentioned that sulfate involved in NPE was usually under the form of sulfuric acid, especially during the initial steps of the growth (Brock et al., 2003; Zhang et al., ~~2004~~2004a; Yue et al., 2010; Bzdek et al., 2012). However, northern California contains very large agricultural regions with a lot of sources of ammonia, which could possibly neutralize sulfate in the ultrafine mode. Using high mass resolution mass spectra acquired under PToF mode, we determined the size distributions of ammonium and sulfate based on those of the NH_3^+ and SO^+, which are the ions of ammonium and sulfate, respectively, with the highest signal-to-noise ratio (see supplementary material for details of this data treatment). As shown in Fig. ~~9~~10, despite relatively noisy data, the size distributions suggest that sulfate was fully neutralized by ammonium in the entire size range, including ultrafine particles. Moreover, we did not observe any difference in the sulfate neutralization between NPE and non-NPE days or between different times of the day. These results indicate that sulfate in ultrafine particles was present in the form of ammonium sulfate and that sulfuric acid was quickly neutralized after condensation.

3.3 Anthropogenic influence on new particle growth events

The average concentrations and diurnal patterns of VOCs, trace gases (O_3, CO, NO_x), BC, and meteorological parameters (temperature, relative humidity, and solar radiation) during NPE days and non-event days were compared (Fig. ~~10~~11, Fig. S3 and S4, and Table 2). An important difference between NPE and non-event days was the concentrations of photo-oxidation products (formaldehyde and acetaldehyde) and anthropogenic precursors (BC, CO and toluene), which were all significantly higher during NPE days than during non-event days. Photo-oxidation products were on average ~50% more concentrated on NPE days (formaldehyde: 2.71 ± 1.39 ppb vs. 1.83 ± 0.81 ppb during non-NPE days; acetaldehyde: 0.97 ± 0.47 ppb vs. 0.71 ± 0.24 ppb). The sum of methacrolein (MACR) and methyl vinyl ketone (MVK), which are the first generational products of isoprene oxidation, was also ~20% higher during NPE days: 0.98 ± 0.79 ppb vs. 0.75 ± 0.50 ppb. These ~~results suggest that the condensation~~markers of ~~these compounds, and~~oxidation are likely correlated with other semi-volatile compounds co-generated during photo-oxidation, which could condense onto the surface of particles and could be an important factor driving the growth of new particles. Moreover, the diurnal patterns of these compounds during NPE and non-NPE days show a clear difference during the afternoon, whereas ~~these~~the differences are much smaller during nighttime (Fig. S4). This result stresses the influence of photochemistry on the formation and growth of new particles.

The average concentrations of isoprene were almost identical during NPE and non-event days (Fig. ~~10). Comparatively,~~11) but the enhancements of anthropogenic species during NPE days were more dramatic. The average concentrations of BC (0.042 ± 0.028 µg m^{-3} during ~~NPG~~NPE days vs. 0.027 ± 0.017 µg m^{-3} during non-NPE days), CO (130 ± 27.0 vs. 99.8 ± 19.8 ppb), NO_x (3.8 ± 3.3 vs. 2.7 ± 3.5 ppb), HOA (0.16 ± 0.15 vs. 0.11 ± 0.08 µg m^{-3}), and toluene (0.060 ± 0.037 vs. 0.038 ± 0.019 ppb; Table 2) were ~~significantly (30 – 60%) higher on NPE days.~~30-60% higher on NPE days. According to the Student's t-test, the difference between NPE and non-NPE days was significant (i.e., $p<0.05$) for all the anthropogenic species, except for NO_x. The ozone concentrations, however, were very similar between two types of days (46.2 ± 10.5 ppb during non-~~NPG~~NPE vs. 43.5 ± 14.2 ppb). These results point out the importance of the anthropogenic influence on the formation and growth of new particles~~, most of these events occurring in the urban plume from Sacramento.~~. However, during a study undertaken at the Blodgett Forest, which is located ~35 km on the northeast of the present sampling site and ~75 km downwind from Sacramento, Lunden et al. (2006) observed new particle growth events when the degree of anthropogenic influence was significantly reduced.

~~It is interesting to note that the~~The relative humidity (RH) was ~~on average 18%~~ higher on NPE days (45 ± 13 %) compared to non-~~event~~NPE days (27 ± 12 %). Previous studies, however, found contradictory links between ~~new particle growth events~~NPEs and RH. For example, Lunden et al. (2006) and Charron et al. (2007) observed ~~growth events when~~much higher RH ~~was high, while non-event~~during NPEs days ~~were characterized by significantly lower RH.~~than non-NPE days. In addition, most of the previous studies reported ~~NPE~~NPEs when the RH was low (Boy and Kulmala, 2002; Hamed et al., 2007; Jeong et al., 2010; Hamed et al., 2011; Guo et al., 2012). The exact role of RH in ~~new particle formation and growth~~NPEs is not clearly elucidated yet. According to Hamed et al. (2011)~~, who used a combination of field data, theoretical calculations and numerical models,~~, the anti-correlation between RH and ~~new particle growth~~NPEs would simply be due to the fact that solar radiation and photochemistry usually peak at noon when the RH exhibits its lower value. However, in our case, this does not seem to ~~have influenced~~explain the ~~growth events of the present study~~different behavior of RH between NPE and non-NPE days, since the weather was sunny during the entire field campaign.~~ The only point~~ A possible reason is that the RH was much lower during northwesterly wind periods (Setyan et al., 2012), during which we usually did not observe ~~growth events~~NPEs.

Fig. ~~11~~12 shows the average size-resolved mass spectra of organics in 40-120 nm (D_{va}) particles during NPE days and non-event days, along with the mass spectra of biogenic SOA and urban transport SOA reported in Setyan et al. (2012). The average mass spectrum of organics before the growth (i.e., between 8:00 and 10:00) was subtracted in order to remove the influence of particles existing before the start of the growth events. Therefore, the spectra shown in Fig. ~~11~~12 are the average mass spectra of organic matter that contributed to the growth of 40-120 nm particles between 10:00 – 16:00 during NPE days ($\Delta Org_{40\text{-}120nm}^{NPE}$) and during non-event days ($\Delta Org_{40\text{-}120nm}^{non\text{-}NPE}$), respectively. As shown in Fig. ~~11a~~12a, the spectrum of $\Delta Org_{40\text{-}120nm}^{NPE}$ is dominated by the signal at *m/z* 44 (mostly CO_2^+), while that of *m/z* 43 (mostly $C_2H_3O^+$) is approximately the half of it. The spectrum of $\Delta Org_{40\text{-}120nm}^{NPE}$ is very similar to that of urban transport SOA ($r^2 = 0.95$; Fig. ~~11b~~12b) but its correlation coefficient towards the spectrum of biogenic SOA is lower ($r^2 = 0.87$). On the other hand, the spectrum of $\Delta Org_{40\text{-}120nm}^{non\text{-}NPE}$ is very similar to that of biogenic SOA, as shown by the scatterplot of Fig. ~~11d~~12d. We further performed multilinear regression analyses to represent the mass spectra of $\Delta Org_{40\text{-}120nm}^{NPE}$ and $\Delta Org_{40\text{-}120nm}^{non\text{-}NPE}$, respectively, as the linear combinations of the spectra of urban transport SOA and biogenic SOA. Based on this analysis, we estimated that during NPE days, ~ 74% of the organic mass that contributed to the growth of ultrafine particles was SOA formed in urban transport plumes. During non-event days, the growth of ultrafine mode organics, which was much slower compared during NPE, was primarily (~ 76% by mass) due to SOA influenced by regional biogenic emissions.

These results, coupled to the higher concentrations of anthropogenic compounds on NPE days suggest that the growth of new particles in the Sierra Nevada Foothills was mainly driven by anthropogenic precursors transported from Sacramento and that the growth was likely promoted by the interaction between urban plumes and biogenic emissions. These observations may have important implications in our understanding of SOA formation. For example, models used to assess global SOA budget tend to underpredict the SOA concentrations. However, in a recent study, Spracklen et al. (2011) used a model to estimate the global OA source, and compared their results with worldwide AMS observations. When they took into account anthropogenically-controlled biogenic SOA formation in their estimation of the global OA budget, it reduced considerably the bias between their model and AMS observations.

4 Conclusions

New particle growth events were frequently observed during the US DOE's CARES campaign in northern California in June 2010. Presented here is a description of these events observed with two ~~SMPSs~~MPSSs deployed at Sacramento (T0, urban site) and Cool (T1, rural site at the Sierra foothills). Our results showed that these growth events took place on a regional scale, predominantly during periods of southwestern flow that transports urban plumes and anthropogenic emissions from the Sacramento metropolitan area and the San Francisco Bay Area near Carquinez Strait. Growth rates were on average higher at T0 (7.1±2.7 nm/hr) than at T1 (6.2±2.5 nm/hr), likely due to higher anthropogenic influences at T0. The evolution of the size-resolved chemical composition of these newly formed particles has been investigated in detail with a HR-ToF-AMS deployed at T1. Our results indicate that the new particle growth was mainly driven by organics, with a small contribution of ammonium sulfate. For example, the average increase of the organic mass in ultrafine particles (40-120 nm in D_{va}, which corresponds to 30-85 nm in Stokes (volume equivalent) diameter, assuming no internal voids, sphericity = 1, and density = 1.4 g/cm^3) was 0.7 µg/m^3 during this period, approximately 7 times higher than that of sulfate (0.1 µg/m^3). Our results also indicate that amines were enhanced significantly during the new particle growth, suggesting that this class of compounds likely played a role. The size-resolved mass spectra of organics in the size range 40-120 nm (in D_{va}) during the growth events were very similar to the mass spectrum of anthropogenically-influenced SOA from urban plume. In addition, during the NPE days, the concentrations of photo-oxidation products (formaldehyde, acetaldehyde, sum of methacrolein and methyl vinyl ketone) and species representative of urban emissions (e.g., BC, CO, NO_x, HOA, and toluene) were on average 50% higher than during non-event days. These results suggest that the new particle growth events were mainly driven by the transported urban plumes and that the growth of new particles was enhanced by the interactions between biogenic emissions and transported urban plumes.

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Table 1. Summary of the characteristics of new particle growth events observed at Sacramento (T0) and Cool (T1) in northern California.

Day	T0				T1						
	Growth rate nm/hr	ΔN/Δt #/cm^3/hr	ΔN$_{12\text{-}20\ nm}$ #/cm^3	NPE Event	Growth rate nm/hr	ΔN/Δt #/cm^3/hr	ΔN$_{12\text{-}20\ nm}$ #/cm^3	ΔOrg $_{40\text{-}120\ nm}$ μg/m^3/hr	ΔSO$_4^{2-}$ $_{40\text{-}120\ nm}$ μg/m^3/hr	Wind	NPE Event
6/2/2010	Incomplete SMPS data			N/A[a]	Incomplete SMPS data			0.75	0.10	T0→T1[b]	N/A
6/3/2010	10.5	6.12E+03	5.43E+03	strong	4.1	2.56E+03	3.35E+03	0.98	0.32	T0→T1	stro
6/4/2010	5.4	6.80E+03	5.44E+03	strong	12.5	6.14E+03	1.70E+03	Incomplete PToF data		T0→T1	stro
6/5/2010	10.9	9.57E+03	3.77E+03	strong	9.3	1.07E+03	1.10E+03	No PToF data		T0→T1	wea
6/6/2010	12.1	7.40E+03	4.57E+03	strong	8.8	2.98E+03	1.01E+03	Incomplete PToF data		T0→T1	wea
6/7/2010	10.1	6.40E+03	4.83E+03	strong	7.6	5.86E+03	1.51E+03	0.28	0.063	T0→T1	strong
6/8/2010	4.1	5.64E+03	4.28E+03	strong	7.7	4.19E+03	2.63E+03	0.35	0.075	T0→T1	strong
6/9/2010	7.3	1.21E+04	1.18E+04	strong	6.1	6.92E+03	4.86E+03	0.49	0.075	T0→T1	strong
6/10/2010	6.5	7.76E+03	1.44E+03	weak	4.2	4.31E+03	1.33E+03	0.19	0.0	NW	weak
6/11/2010	7.1	2.48E+03	1.16E+03	weak	-[c]	-	-	-	-	NW	no N
6/12/2010	-	-	-	no NPG	Incomplete data					NW	N/A
6/13/2010	-	-	-	no NPG	-	-	-	-	-	NW	no N
6/14/2010	4.6	1.26E+04	8.67E+03	strong	Incomplete data					T0→T1	N/A
6/15/2010	4.4	6.75E+03	8.81E+03	strong	3.8	4.52E+03	3.29E+03	0.27	0.095	T0→T1	stro
6/16/2010	2.5	4.50E+03	6.55E+03	strong	3.6	1.59E+03	-1.35E+03	-	-	NW	wea
6/17/2010	-	-	-	no NPG	2.9	2.28E+03	8.08E+02	0.19	0.0	NW	wea
6/18/2010	6.7	3.56E+04	8.25E+03	strong	4.3	5.30E+03	1.74E+03	0.32	0.032	T0→T1	stro
6/19/2010	3.4	2.60E+04	9.55E+03	strong	5.9	5.20E+03	1.48E+03	0.19	0.041	T0→T1	wea
6/20/2010	4.1	8.69E+03	6.02E+03	strong	4.4	2.04E+03	-8.86E+02	-	-	NW	wea
6/21/2010	5.2	4.19E+03	1.25E+03	weak	9.5	1.96E+03	9.89E+02	0.17	0.0	NW	wea
6/22/2010	7.6	1.51E+04	5.03E+03	strong	Incomplete SMPS data			0.10	0.0	T0→T1	N/A
6/23/2010	11.1	7.28E+03	1.45E+03	weak	Incomplete SMPS data			0.17	0.060	T0→T1	N/A
6/24/2010	6.4	7.11E+03	8.74E+03	strong	7.6	8.28E+03	2.27E+03	0.64	0.13	T0→T1	stro
6/25/2010	8.0	4.07E+03	4.16E+03	strong	4.7	3.15E+03	9.77E+02	0.31	0.0	T0→T1	weak
6/26/2010	7.7	9.13E+03	6.93E+03	strong	5.3	1.81E+03	8.26E+02	0.27	0.11	T0→T1	weak
6/27/2010	9.3	5.25E+03	6.70E+03	strong	5.6	1.34E+03	1.01E+03	0.11	0.0	T0→T1	weak
6/28/2010				undefined[d]			-	-	-	T0→T1	undefi
mean	7.1	9.57E+03	-5.67E+03	-	6.2	3.76E+03	1.74E+03	0.34	0.06		
std dev	2.7	7.62E+03	2.90E+03		2.5	2.09E+03	1.09E+03	0.24	0.08		
median	6.9	7.20E+03	5.44E+03		~~5.6~~ 5.6	3.15E+03	1.35E+03	0.27	0.06		
min	2.5	2.48E+03	1.16E+03		2.9	1.07E+03	8.08E+02	0.10	0.0		
max	12.1	3.56E+04	1.18E+04		12.5	8.28E+03	4.86E+03	0.98	0.32		

[a] "N/A" stands for not applicable

[b] "T0 → T1" stands for T0 to T1 transport periods

[c] "-" means that no increase was observed

[d] "undefined" means that the ~~SMPS~~MPSS data did not allow to determine whether a growth event took place or not, because of a change in the wind direction during the day.

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Table 2. Summary of average value ± 1 standard deviation for meteorological parameters, particle phase species, and gaseous species during new particle event (NPE) and non-NPE days at the T1 site between 8:00 and 18:00 PDT.

Parameter	NPE days	Non-NPE days
Meteorological data		
Temperature (^oC)	24.2 ± 4.4	25.0 ± 4.1
Relative humidity (%)	45.3 ± 12.6	27.1 ± 12.1
Solar radiation (W m^{-2})	702.9 ± 246.1	792.7 ± 200.4
Particle phase		
Particle number (# cm^{-3})	9.4E3 ± 6.1E3	4.1E3 ± 1.9E3
Growth rate (nm/hr)	6.2 ± 2.5	-
Biogenic SOA (μg m^{-3})	0.90 ± 0.65	0.56 ± 0.27
Urban transport SOA (μg m^{-3})	1.2 ± 0.90	0.54 ± 0.44
HOA (μg m^{-3})	0.16 ± 0.15	0.11 ± 0.08
SO_4^{2-} (μg m^{-3})	0.39 ± 0.22	0.14 ± 0.10
NO_3^- (μg m^{-3})	0.13 ± 0.08	0.054 ± 0.036
BC (μg m^{-3})	0.042 ± 0.028	0.027 ± 0.017
Trace gases (ppb)		
Terpenes	0.058 ± 0.088	0.043 ± 0.034
Isoprene	1.40 ± 1.02	1.35 ± 0.80
MACR + MVK	0.98 ± 0.79	0.75 ± 0.50
Methanol	6.36 ± 3.12	5.36 ± 1.76
Acetone	1.90 ± 1.09	1.64 ± 0.42
Formaldehyde	2.71 ± 1.39	1.83 ± 0.81
Acetaldehyde	0.97 ± 0.47	0.71 ± 0.24
Acetic acid	0.98 ± 1.10	0.87 ± 0.43
Acetonitrile	0.18 ± 0.03	0.17 ± 0.02
Benzene	0.036 ± 0.029	0.031 ± 0.014
Toluene	0.060 ± 0.037	0.038 ± 0.019
O_3	43.5 ± 14.2	46.2 ± 10.5
NO_x	3.8 ± 3.3	2.7 ± 3.5
CO	130.1 ± 27.0	99.8 ± 19.8

Figure 1. Time series of (a, d) wind direction colored by wind speed, (b, e) broadband solar radiation, temperature and relative humidity, and (c, f) particle size distributions at the T0 and T1 sites.

~~**Figure 2.**~~

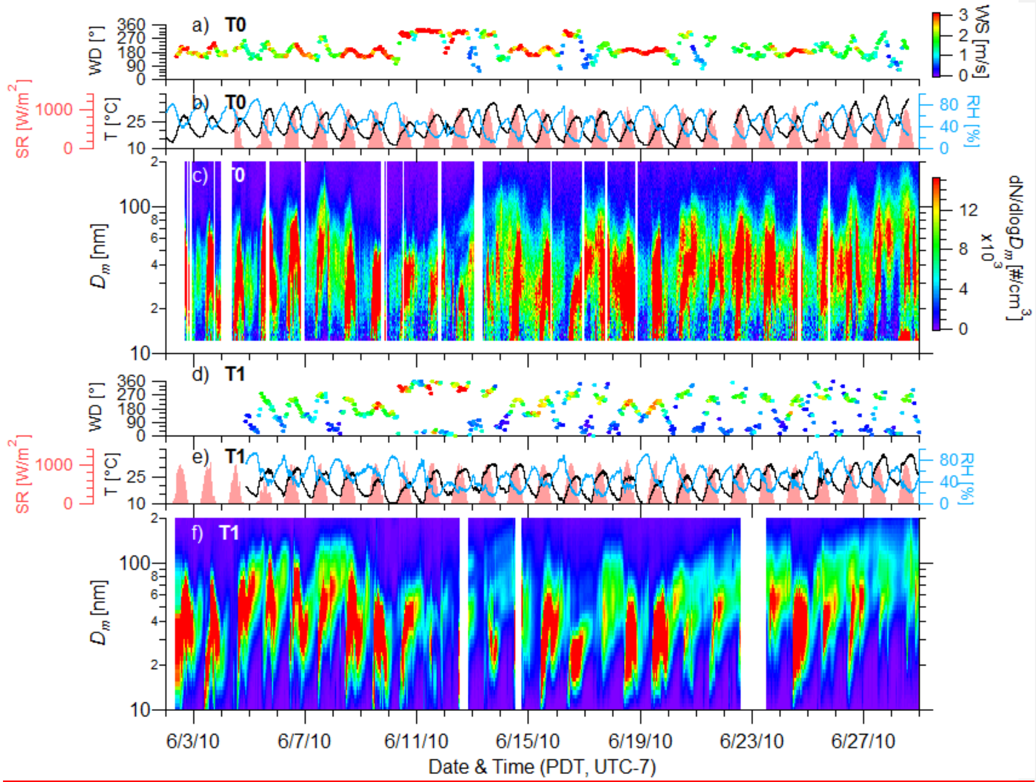

Figure 2. Diurnal patterns of particle number concentrations measured at the T0 and T1 sites during NPE days.

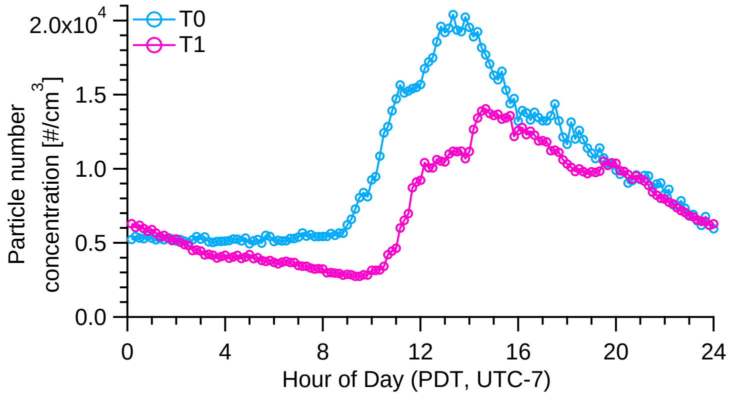

Figure 3. Comparison of the time evolution of the particle size distributions at the (a) T0 and (b) T1 sites on June 26, along with the hourly averaged wind direction (length of the arrows is proportional to the wind speed) for each site. Time series of (c) NR-PM$_1$ species and BC, and (d) three different OA factors.

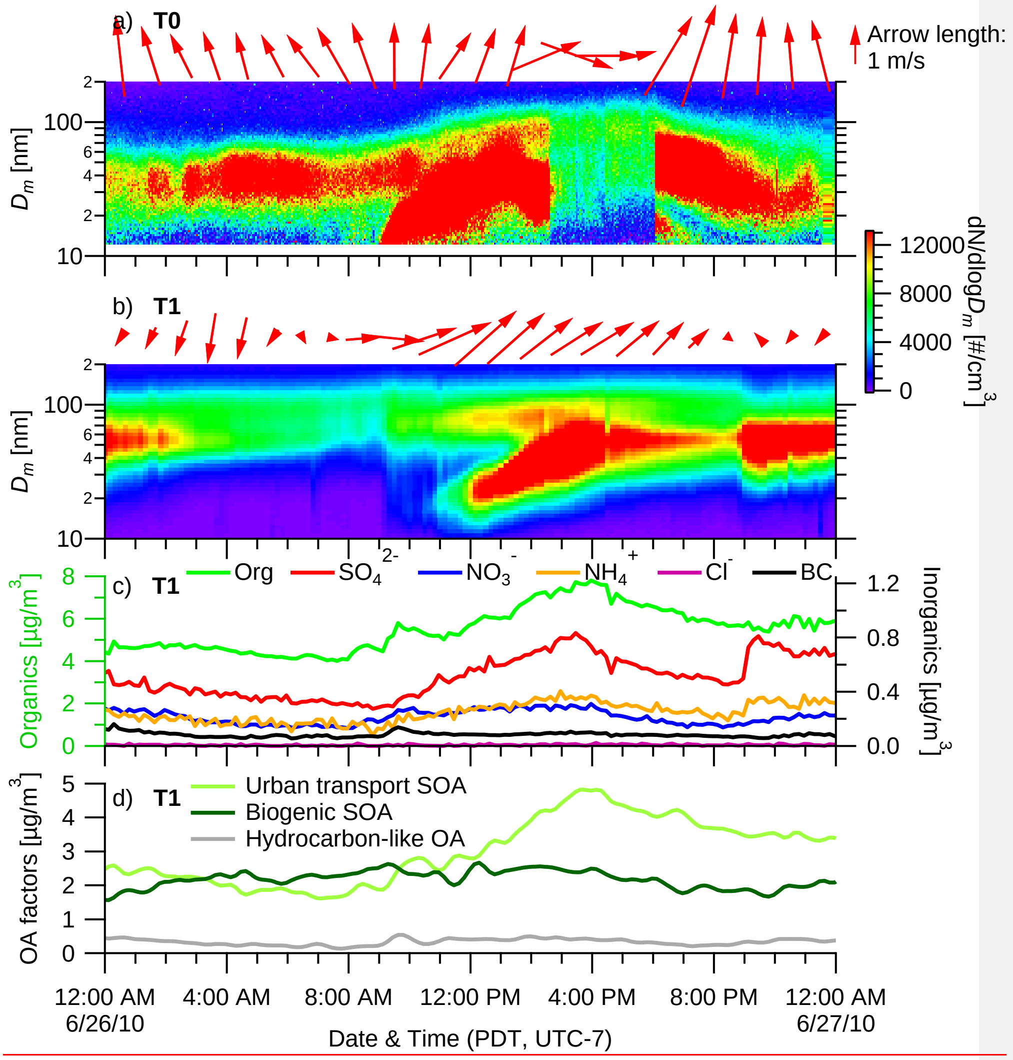

Figure ~~3~~4. Comparisons of the average particle number size distributions for each hour at T0 and T1 during June 26.

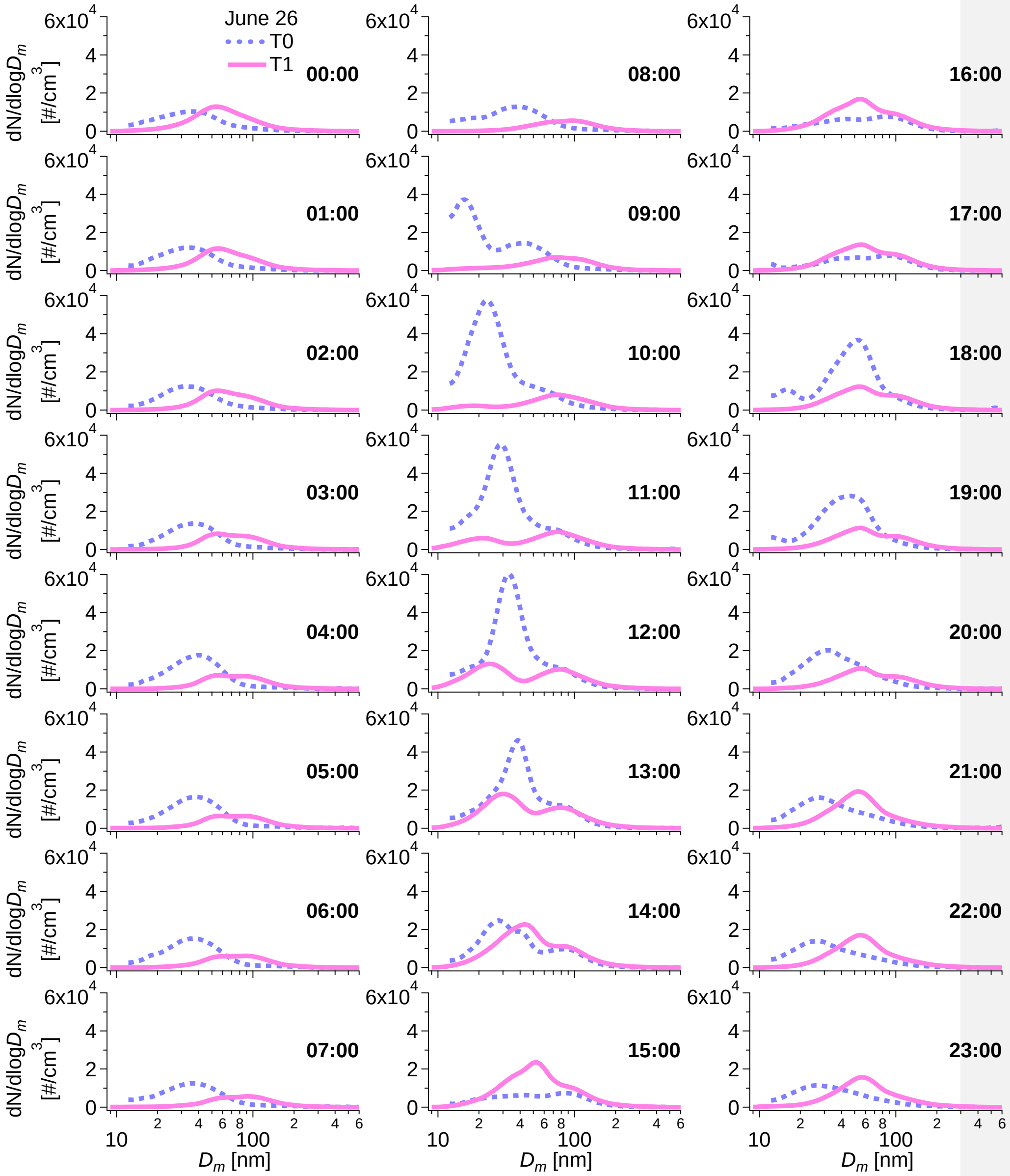

Figure 45. Diurnal size distributions of the particle number concentration at the (a, b) T0 and (c, d) T1 sites during NPE days (left panel) and non-NPE days (right panel). Black crosses correspond to the ~~modes~~modal diameters fitted by log-normal distributions.

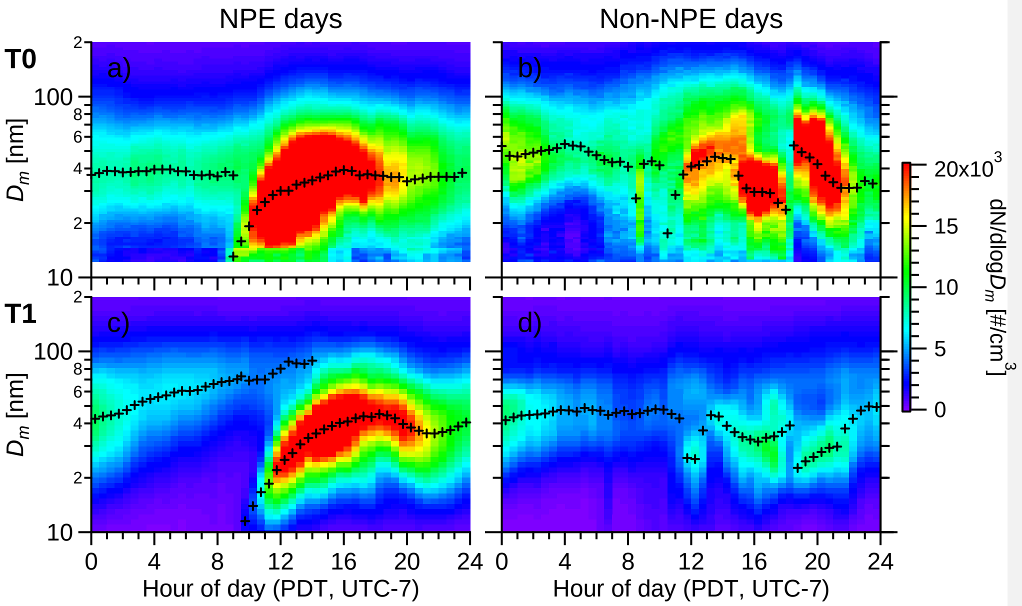

Figure ~~5~~6. Diurnal size distributions of (a, b) Org, (c, d) SO_4^{2-}, and (e, f) particle volume concentrations, and (g, h) daytime wind rose plots (8:00-20:00 PDT) for NPE days (left panel) and non-NPE days (right panel).

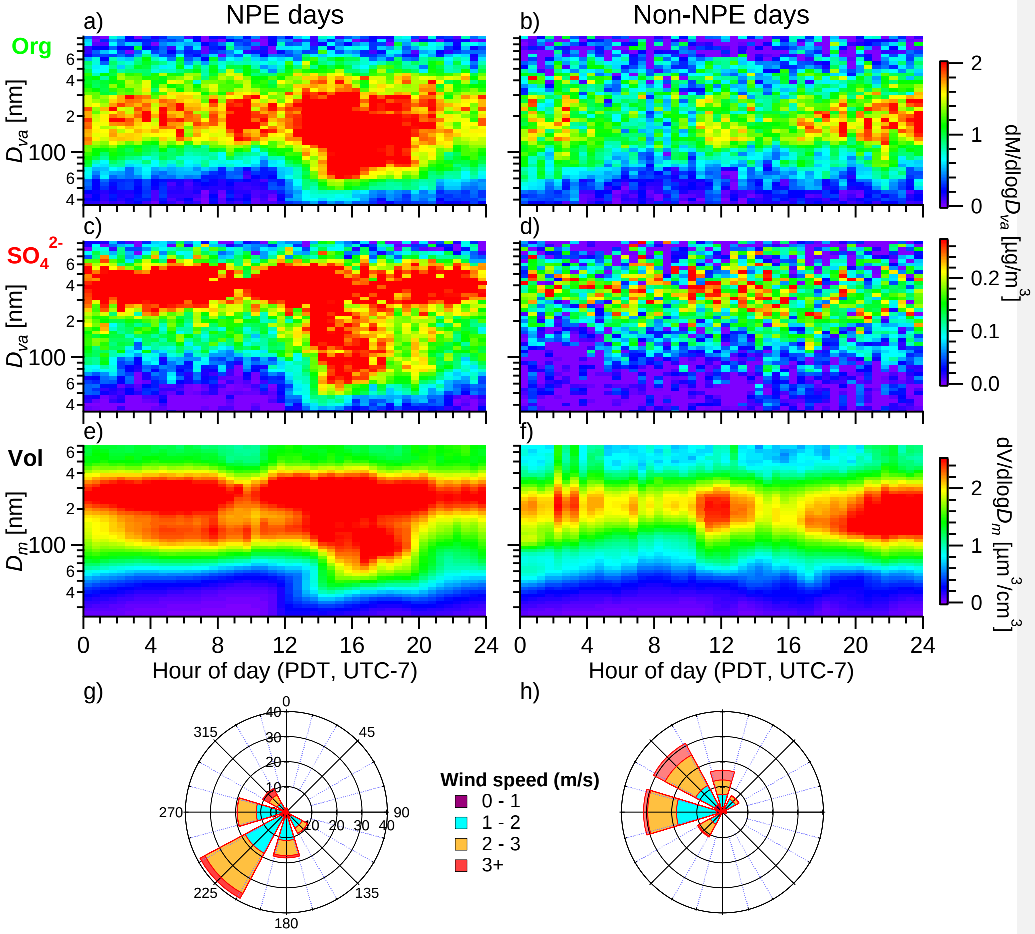

Figure ~~6~~7. Diurnal patterns of the concentrations of (a, b) Org and (c, d) SO_4^{2-} (black circles and lines, right y-axes) and the mass fractions in the range 40-120, 120-200 and 200-800 nm (in D_{va}, left y-axes) during NPE days (left panel) and non-NPE days (right panel).

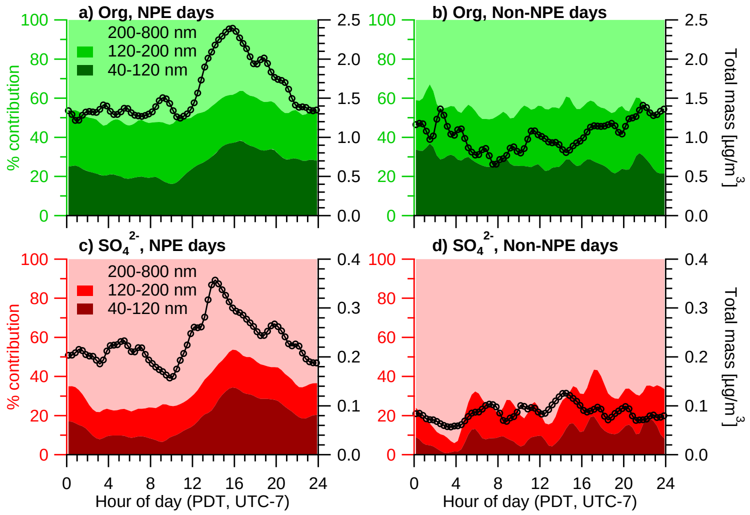

Figure ~~7~~8. Diurnal patterns of (a) particle number concentration (10-15 nm), (b) urban transport SOA, (c) biogenic SOA, (d) SO_4^{2-} (40-120 nm in D_{va}), (e) Org (40-120 nm in D_{va}), and (f) N-containing organic ions (= CHN^+ + CH_4N^+ + $C_2H_3N^+$ + $C_2H_4N^+$) and ammonium during NPE (solid symbols) and non-NPE (open symbols) days.

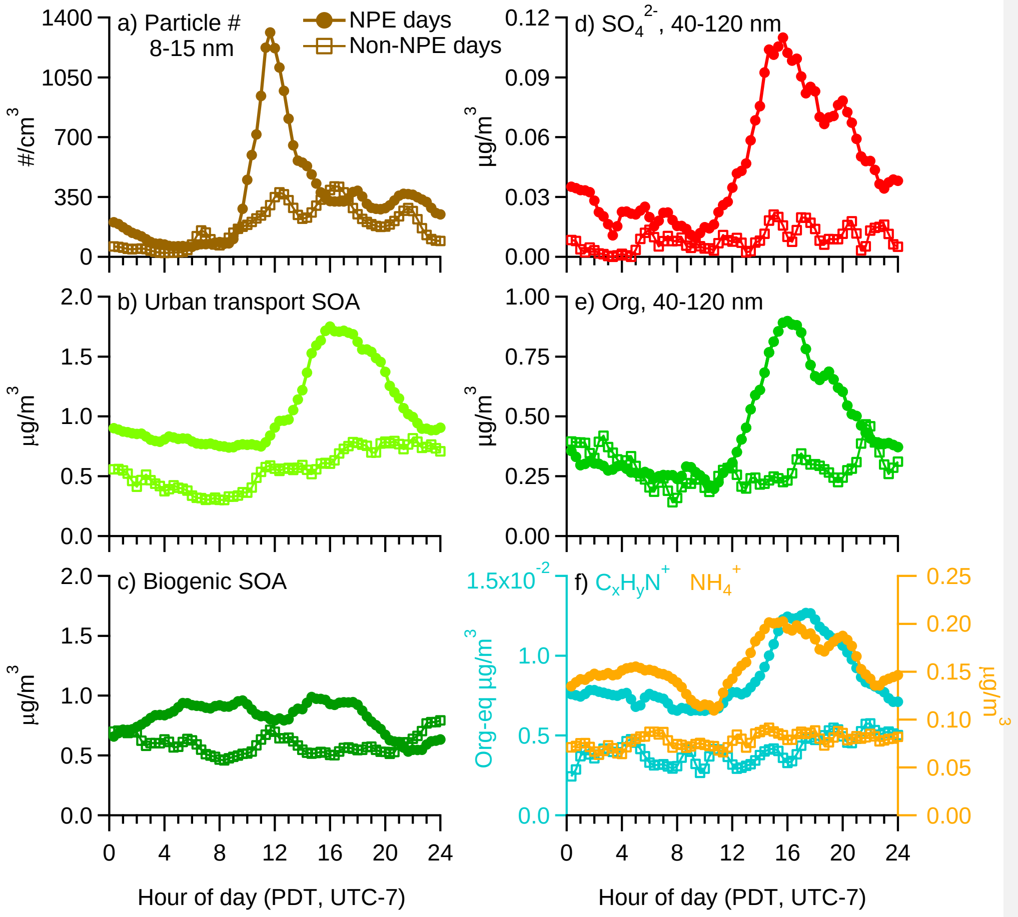

Figure ~~8~~9. 2-hour averaged size distributions of (a, b) particle number ~~(a, b)~~ and (c, d) volume ~~(c, d), SO$_4^{2-}$~~, (e, f~~),~~) SO$_4^{2-}$, and ~~Org~~ (g, h) Org during NPE days (left panel) and non-NPE days (right panel) between 8:00 and 18:00 (PDT).

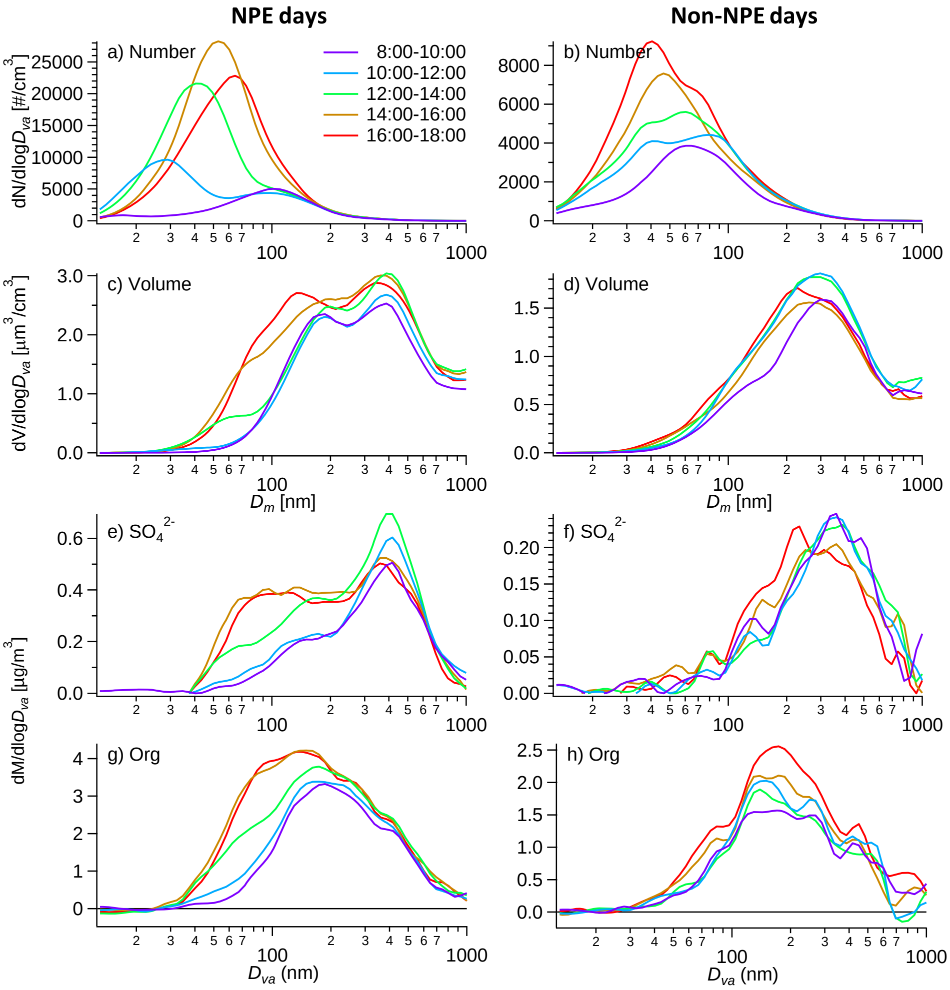

Figure ~~9~~10. Size distributions of SO_4^{2-}, NH_4^+ and the ratio of measured NH_4^+ to predicted NH_4^+ (= 2·SO_4^{2-}~~/48×~~·18/96) between ~~6:00-7:00~~ (a~~, b),~~) 10:00-11:00~~ (c, d),~~, (b) 14:00-15:00 ~~(e, f),~~, and (c) 18:00-19:00 ~~(g, h)~~ during ~~new particle event (~~NPE~~; left panels) and non-NPE (right panels)~~ days.

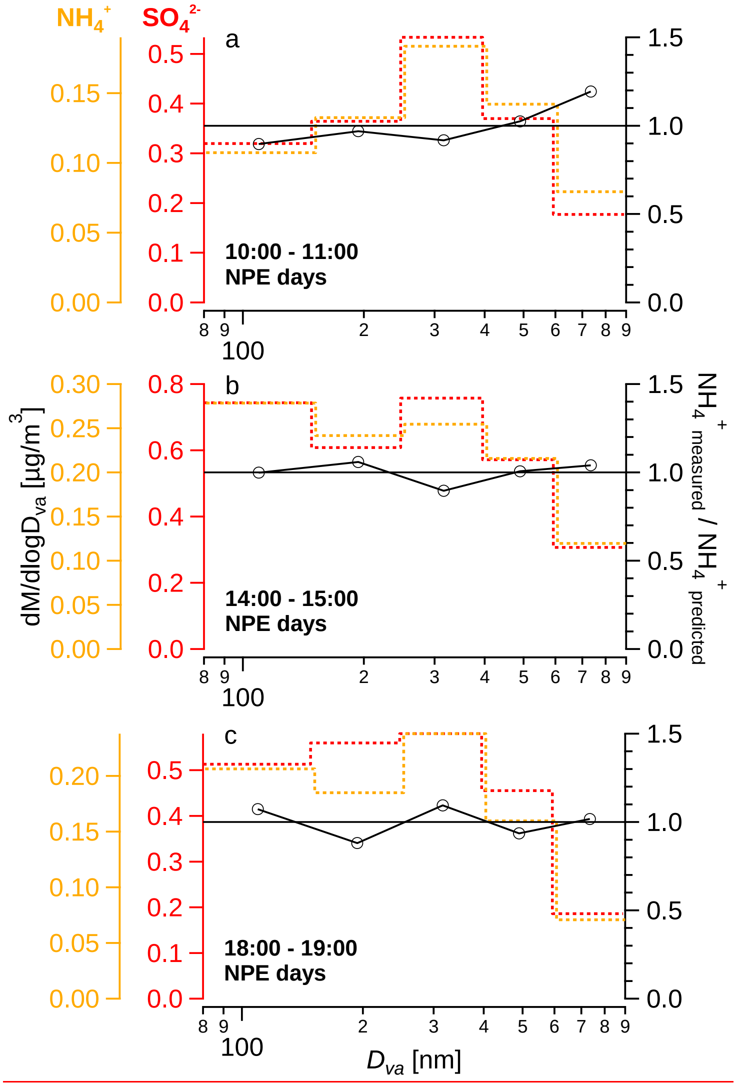

Figure ~~10~~11. (b) Average concentrations of VOCs, O_3, NO_x, CO, BC, NR-PM_1 species, different OA factors, and N-containing organic ions (= $CHN^+ + CH_4N^+ + C_2H_3N^+ + C_2H_4N^+$) between 8:00 and 18:00 (PDT) during NPE and ~~Non~~non-NPE days. (a) NPE days / Non-NPE days ratios for the same parameters.

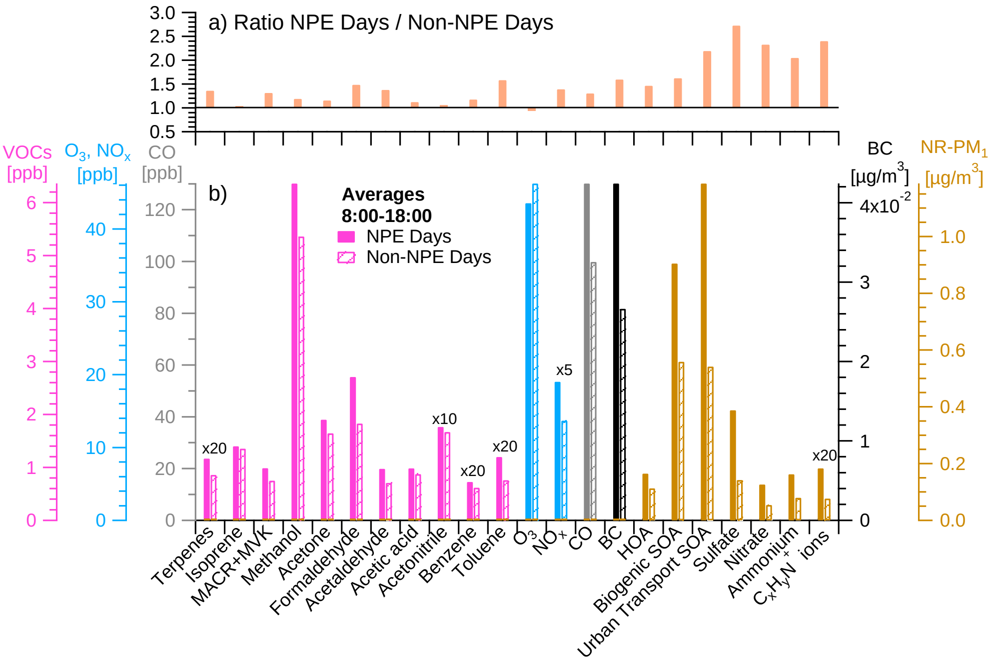

Figure ~~11~~12. Average mass spectra of (a) urban transport SOA and $\Delta Org_{40\text{-}120nm}$ (i.e., organics that contribute to the growth of 40-120 nm particles) during NPE days, and (c) biogenic SOA and $\Delta Org_{40\text{-}120nm}$ during Non-NPE days. Scatterplots that compare the mass spectra of (b) urban transport SOA vs. $\Delta Org_{40\text{-}120nm}$ during NPE days, and (d) biogenic SOA vs. $\Delta Org_{40\text{-}120nm}$ during non-NPE days. The data fitting of these two scatterplots was performed using the orthogonal distance regression (ODR).