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Wintertime particulate pollution episodes in an urban valley of the Western US: a case study

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Abstract. This study investigates the causes of elevated $PM_{2.5}$ episodes and potential exceedences of the US National Ambient Air Quality Standards (NAAQS) in Truckee Meadows, Nevada, an urban valley of the Western US, during winter 2009/2010, an unusually cold and snowy winter. Continuous $PM_{2.5}$ mass and time-integrated chemical speciation data were acquired from a central valley monitoring site, along with meteorological measurements from nearby sites. All nine days with $PM_{2.5} > 35 \,\mu g \, m^{-3}$ showed 24-h average temperature inversion of 1.5–4.5 °C and snow cover of 8–18 cm. Stagnant atmospheric conditions limited wind ventilation while highly reflective snow cover reduced daytime surface heating creating persistent inversion. Elevated ammonium nitrate (NH_4NO_3) and water associated with it are found to be main reasons for the $PM_{2.5}$ exceedances. An effective-variance chemical mass balance (EV-CMB) receptor model using locally-derived geological profiles and inorganic/organic markers confirmed secondary NH_4NO_3 (27–37 %), residential wood combustion (RWC; 11–51 %), and diesel engine exhaust (7–22 %) as the dominant $PM_{2.5}$ contributors. Paved road dust and de-icing materials were minor, but detectable contributors. RWC is a more important source than diesel for organic carbon (OC), but vice versa for elemental carbon (EC). A majority of secondary NH_4NO_3 is also attributed to RWC and diesel engines (including snow removal equipment) through oxides of nitrogen (NO_x) emissions from these sources. Findings from this study may apply to similar situations experienced by other urban valleys.

1 Introduction

The Truckee Meadows is a mountain valley in Northern Nevada of the US with the Sierra Nevadas to its west and the Great Basin to its east. The area of the valley floor is $\sim 25 \, km^2$, containing downtowns of the cities of Reno and Sparks with a population of $\sim 200\,000$ (2010 census). It is distant from major metropolitan areas by at least 200 km. Similar urban valleys are common in the Western Cordillera region, including Salt Lake City, Utah, Boise-Nampa, Idaho, Missoula, Montana, and Kelowna, British Columbia (Canada). Air quality in such valleys is determined by the interaction between synoptic winds and local thermal circulation (Fernando, 2010). Under stable conditions, pollutants released from the valley floor tend to be confined by surrounding mountain ranges leading to elevated pollution episodes.

Particulate matter 2.5 ($PM_{2.5}$, airborne particles with aerodynamic diameter equal or less than 2.5 µm) is a criteria air pollutant regulated by the US Environmental Protection Agency (USEPA) for its adverse effects on public health, visibility, and environment (USEPA, 2004). $PM_{2.5}$ episodes often show patterns (e.g., chemical composition and seasonality) related to region-specific meteorology and sources (Chu, 2004; Tai et al., 2010). Compliance monitoring for $PM_{2.5}$ in the Truckee Meadows (Mendoza and Inouye, 2011) showed that most of the high values occurred during winter, with some excursions during late summer and fall when smoke from regional wildfires was a contributor. Ling-Barnes (2010) estimates that most primary $PM_{2.5}$ emissions

in the Truckee Meadows are from area (non-point) sources associated with wintertime residential heating. Other important sources include motor vehicle engine exhaust and fugitive dust.

Although Mendoza and Inouye (2011) have demonstrated attainment of PM$_{2.5}$ National Ambient Air Quality Standards (NAAQS) of 15 µg m^{-3} annual arithmetic mean, averaged over three years, and 35 µg m^{-3} for the 98th percentile of 24-h averages, averaged over three years (Bachmann, 2007; Chow et al., 2007b), in the Truckee Meadows since 2001, several 24-h PM$_{2.5}$ concentrations exceeding 35 µg m^{-3} were observed from 2008 to 2010. Considering also that USEPA (2012) has recently proposed reducing the annual NAAQS to 12–13 µg m^{-3}, these high episodes raise concerns of potential NAAQS non-attainment in the future.

This study investigates interactions between meteorology and emissions that lead to wintertime PM$_{2.5}$ episodes in the Truckee Meadows valley. The analysis is based on regulatory monitoring data and additional measurements on archived samples that facilitate source apportionment using a chemical mass balance (CMB) approach. Findings from this study provide information for developing strategies to reduce PM$_{2.5}$ levels in this and similar urban valleys.

2　Ambient monitoring data collection and analysis

The main PM$_{2.5}$ monitoring site in Reno (RENO_C: 39.525079° N, −119.807723° W, 1371 m a.m.s.l.) is located in a residential/commercial area south of the Truckee River, ∼ 2 km west of the I-580 north/south freeway and ∼ 1 km south of the I-80 east/west freeway. A Mexican restaurant is located ∼ 20 m east of the station. As part of the USEPA Speciation Trends Network (STN), PM$_{2.5}$ filters have been acquired at RENO_C every 3rd day with speciation samplers (Met One SASS + URG 3000N Carbon Sampler) for chemical characterization (Chow et al., 2010; Flanagan et al., 2006). Additional PM$_{2.5}$ and PM$_{10}$ filter samples are taken with Federal Reference Method (FRM) samplers for gravimetric analysis of mass concentrations. Hourly average PM$_{2.5}$ and oxides of nitrogen (NO$_x$) are also measured by a beta attenuation monitor (BAM; MetOne E-BAM; Grants Pass, OR) and a chemiluminescent analyzer (42C, Thermo Environmental Instruments, Franklin, MA), respectively. The sampling probes are situated ∼ 4 m above ground level and ∼ 1 m above the rooftop of the shelter. For this study, data for the December 2008–January 2009 and December 2009–January 2010 were retrieved from USEPA's Air Quality System database (AQS; http://www.epa.gov/ttn/airs/airsaqs/index.htm). Concurrent meteorological observations were obtained from stations on the valley floor and at higher elevations, as shown in Fig. 1.

Based on the FRM measurements, there were four 24-h PM$_{2.5}$ samples exceeding 35 µg m^{-3} out of 20 measurements at the RENO_C site during December 2009 and Jan-

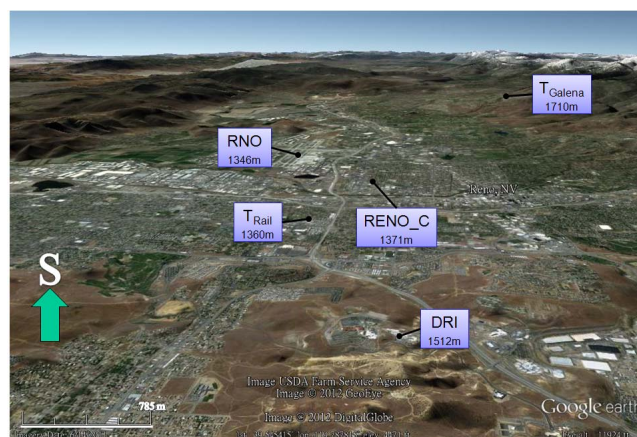**Fig. 1.** Topography of the Truckee Meadows valley and locations/elevations of PM$_{2.5}$ monitoring site (RENO_C), valley-floor weather station (Reno Rail Yard, T_{Rail}), elevated weather station (Galena, T_{Galena}), Reno-Tahoe International Airport (RNO), and the Desert Research Institute (DRI).

uary 2010 (i.e., 9, 15, 18 December 2009, and 5 January 2010). Daily averaged BAM data reported five additional PM$_{2.5}$ exceedances (i.e., 10, 11, 14, 19, and 20 December 2009) during the period. The December 2009–January 2010 average FRM PM$_{2.5}$ mass concentration was 17.9 µg m^{-3}, ∼ 45 % higher than the December 2008–January 2009 average (12.3 µg m^{-3}). PM$_{2.5}$ mass from the BAM and the speciation samplers showed good agreement with the FRM mass ($r^2 > 0.95$ and average differences within ± 7 %) (Watson et al., 2011).

2.1　Meteorological characteristics of PM$_{2.5}$ episodes

Temperature inversions often reflect calm winds and low vertical dispersion that enhance accumulation of pollutants released from the surface. Valley-floor and elevated meteorological observations (Fig. 1) were used to characterize the strength of inversions in the Truckee Meadows area. The Reno Rail Yard site (39.5391° N 119.78605° W) is on the valley floor at an elevation of 1360 m mean sea level (m.s.l.), ∼ 1.3 km northeast of RENO_C, while the Galena Remote Automated Weather Station (RAWS) site (39.3794° N 119.8319° W) is at an elevation of 1710 m m.s.l., generally upwind of the Truckee Meadows, and at the top of the atmospheric boundary layer.

Good relationships were found between PM$_{2.5}$ mass at the RENO_C site and vertical temperature differences. For winter 2009/2010, Fig. 2a shows that there were three periods (i.e., 8–11 December 2009; 14–20 December 2009; and 3–6 January 2010) of persistent, multi-day inversions with maximum inversion strengths ($\Delta T = T_{\text{Galena}} - T_{\text{Rail}}$) > 5 °C and minimum $\Delta T > -2$ °C. During these periods, ΔT peaked at or just before midday. As surface heating increased in the afternoon, the inversion coupled to layers aloft near 16:00 local
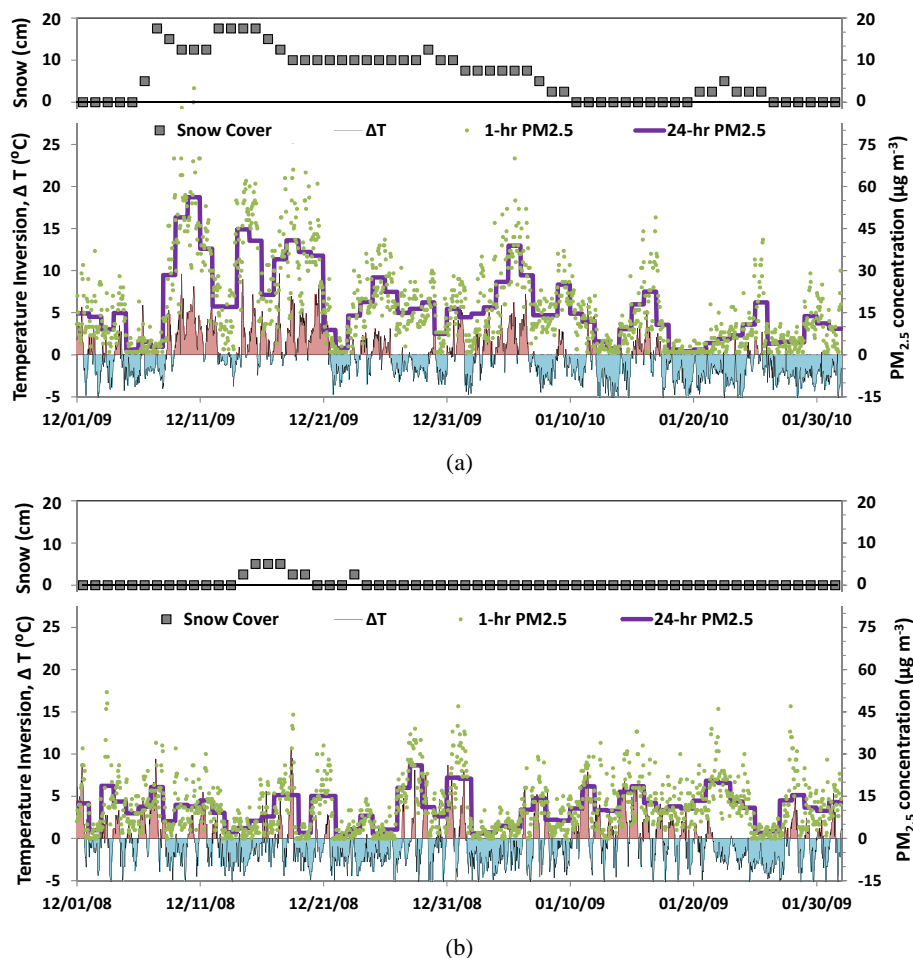
Fig. 2. Snow cover (daily), temperature inversion ($\Delta T = T_{\text{Galena}} - T_{\text{Rail}}$), and corresponding PM$_{2.5}$ mass concentrations by beta attenuation monitor (BAM) during: **(a)** December 2009–January 2010, and **(b)** December 2008–January 2009. $\Delta T > 0$ (i.e., with inversion) and $\Delta T < 0$ are marked by red and blue, respectively.

standard time (LST), but re-instated itself soon after sunset. The highest PM$_{2.5}$ concentrations, including all 24-h exceedances, occurred near the middle of each inversion period. For other times of the winter, inversions were weak and short-lived, typically lasting no more than several hours. The winter also experienced a few snow storms during which no inversions occurred throughout a day.

There were neither extended periods of strong inversions nor PM$_{2.5}$ exceedances during winter 2008/2009 (Fig. 2b). Daily average ΔT is positively correlated with 24-h PM$_{2.5}$ measured by BAM with r^2 of 0.785 and 0.420 for winter 2009/2010 and winter 2008/2009, respectively. Inversions have been confirmed by Silcox et al. (2012) to strongly influence PM$_{2.5}$ concentration in the Salt Lake City valley, though in that study inversion (or stability) was measured through heat deficit, the energy required to dry adiabatically mix a layer.

December 2009 was the sixth coldest and fifth snowiest December (39.6 cm snowfall) recorded since 1931 at the

Reno-Tahoe International Airport (RNO, see Fig. 1). Snowfall was low to moderate for the other three months (1.3–15.0 cm; December 2008, January 2009, and January 2010). Highly reflective and melting snow cover reduces daytime surface heating, which allows the formation of persistent inversions throughout the day under stable synoptic conditions. Forty-eight out of 124 days in the two winter periods recorded snow cover of ≥ 2.5 cm at RNO with an average daily ΔT of 0.1 ± 3.6 °C, which is significantly higher ($p = 0.005$) than the average daily ΔT of -1.6 ± 2.9 °C for the 76 days without snow cover. High ΔT can occur on days without snow cover, but heating in the afternoon on those days is usually strong enough to break the inversion completely, avoiding day-to-day accumulation. This was the case for most of winter 2008/2009 as shown in Fig. 2b. Snow cover also increased relative humidity (RH), as RH averaged 78 % on snow-cover days and 66 % on no-snow-cover days.

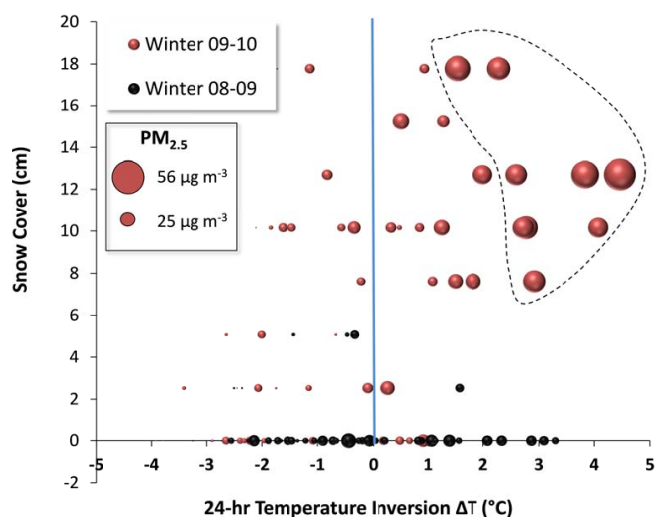Figure 3 compares BAM PM$_{2.5}$ in both winters as a function of ΔT and snow cover. All nine days with

Fig. 3. Average 24-h $PM_{2.5}$ mass concentration (bubble diameter) by beta attenuation monitor (BAM) as a function of temperature inversion ($\Delta T = T_{Galena} - T_{Rail}$) and snow cover. The largest bubble corresponds to $56.1\,\mu g\,m^{-3}$ of $PM_{2.5}$. The dashed circle encloses all nine days that exceeded the USEPA 24-h $PM_{2.5}$ National Ambient Air Quality Standard (NAAQS) of $35\,\mu g\,m^{-3}$.

$PM_{2.5} > 35\,\mu g\,m^{-3}$ experienced 24-h ΔT of 1.5 to 4.5 °C and snow cover of 8–18 cm. There had been much less snowfall during winter 2008/2009 with surface snow cover always < 8 cm. No $PM_{2.5} > 35\,\mu g\,m^{-3}$ were recorded for winter 2008/2009 despite some ΔT that were as high as the winter 2009/2010 exceedance days. On the other hand, days with deep snow cover but no inversion saw only moderate $PM_{2.5}$ levels.

The combination of high 24-h ΔT and snow cover increases the chance of prolonged primary aerosol accumaltion and secondary aerosol (e.g., ammonium nitrate [NH_4NO_3] and secondary organic carbon [SOC]) formation, thus leading to $PM_{2.5}$ episodes. In addition, snow cover necessitates application of de-icing material during snow plowing and blowing. These activities increase the use of diesel and gasoline engines that may emit more primary $PM_{2.5}$ and NO_x (a nitrate [NO_3^-] precursor) than during drier periods. More cold starts for commuters can also increase emissions over those found during warmer weather (Cadle et al., 2001; Chen et al., 2001; Weilenmann et al., 2009; Nam et al., 2010).

2.2 Chemical characteristics of $PM_{2.5}$ episodes

$PM_{2.5}$ mass (speciation mass hereafter unless mentioned otherwise) and speciation data acquired from AQS contain uncertainty estimates for all variables except organic carbon (OC), elemental carbon (EC), and thermal carbon fractions by the IMPROVE_A protocol (Chow et al., 2007a, 2011). In addition to OC and EC, there are 48 elemental concentrations reported along with water soluble ammonium (NH_4^+), NO_3^-, sulfate ($SO_4^=$), sodium (Na^+), and potassium (K^+).

Mass closure for the $PM_{2.5}$ samples was calculated by summing NH_4^+, NO_3^-, $SO_4^=$, organic matter (OM), EC, crustal material, and salt (potentially from de-icing materials) as the reconstructed mass (Malm et al., 1994; Chen et al., 2002), where:

$$OM = 1.4 \times [OC] \tag{1}$$

$$\begin{aligned} Crustal\,Material &= 2.2 \times [Al] + 2.49 \times [Si] + 1.64 \times [Ca] \\ &\quad + 2.42 \times [Fe] + 1.94 \times [Ti] \end{aligned} \tag{2}$$

$$Salt = [Na^+] + [Cl] \tag{3}$$

Figure 4 shows the time series of measured and reconstructed $PM_{2.5}$ mass during the winter 2008/2009 and 2009/2010, while Fig. 5 compares concentrations of major constitutes between the two winter periods. Since $SO_4^=$ levels were low, NH_4^+ was mostly associated with NO_3^- as NH_4NO_3, which, along with OM, dominated the $PM_{2.5}$ mass. The sum of constituents exceeds the measured mass for samples from winter 2008/2009 except 8 December 2008 and 13 January 2009, leading to overall negative unidentified mass (UnID). For winter 2009/2010, however, UnID is generally positive. Negative UnID could result from overestimating OM. Using a multiplier of 1.4 for OC (Eq. 1) may be too high for sources such as gasoline and diesel exhausts (Russell, 2003; El-Zanan et al., 2005; Aiken et al., 2008) and the organic sampling artifact may not be sufficiently subtracted for the STN (Watson et al., 2009; Chow et al., 2010). Positive UnID indicates unaccounted species such as water. Mass closure on 30 December 2009 and 20 January 2010 was particularly poor, raising questions with regard to mass measurements for the two days.

The higher average $PM_{2.5}$ concentration during winter 2009/2010 mainly resulted from larger NH_4NO_3 and UnID contributions (Fig. 5). Despite high OM concentrations on episode days (i.e., 9, 15, 18 December 2009, and 5 January 2010), OM averaged lower for winter 2009/2010 than for winter 2008/2009. As an example, the 5 January 2010 sample contains 3.2 times the NO_3^- concentration of 1 January 2009 ($8.4\,\mu g\,m^{-3}$ versus $2.6\,\mu g\,m^{-3}$) and a higher UnID ($4.6\,\mu g\,m^{-3}$) but with only comparable OM ($17.2\,\mu g\,m^{-3}$ versus $18.5\,\mu g\,m^{-3}$ for 5 January 2010 and 1 January 2009, respectively). The Teflon-membrane filters were weighed at 30–40 % RH, and water strongly bound to NH_4NO_3 might not be removed completely due to a hysteresis effect (Speer et al., 2003). If so, elevated NH_4NO_3 was the main cause of $PM_{2.5}$ exceedances during winter 2009/2010. This is consistent with observations from the nearby, but much larger, San Joaquin Valley, California in winter (Watson and Chow, 2002; Chow et al., 2006; Chen et al., 2007b).

Teflon-membrane filter samples and the remnants of quartz-fiber filter samples for winter 2009/2010 were retrieved from the STN archive and submitted to additional analyses (Chow and Watson, 2012), including: (1) thermal
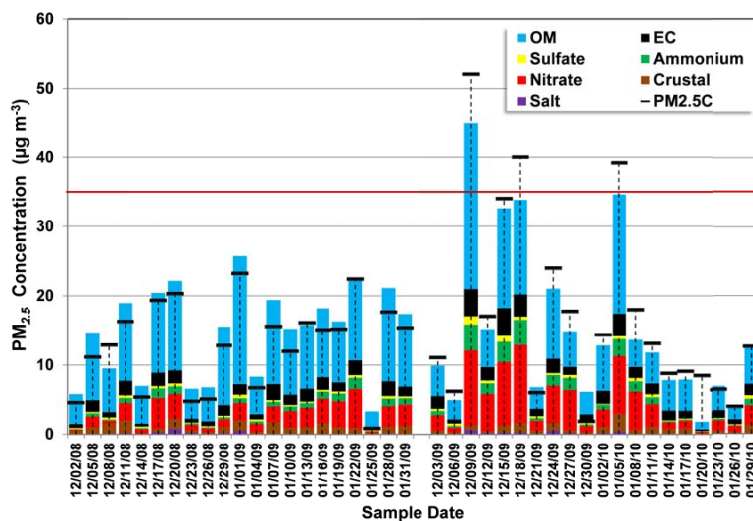
Fig. 4. PM$_{2.5}$ mass reconstruction compared to speciation mass (PM$_{2.5}$C) for winter 2008/2009 and 2009/2010. The red line indicates the USEPA 24-h PM$_{2.5}$ National Ambient Air Quality Standard (NAAQS) of 35 µg m^{-3}. The 15 December 2009 sample exceeds the NAAQS by FRM mass but not by speciation mass (34 µg m^{-3}). OM = 1.4 × [OC]; Crustal Material = 2.2 × [Al] + 2.49 × [Si] + 1.64 × [Ca] + 2.42 × [Fe] + 1.94 × [Ti]; and Salt = [Na^{+}] + [Cl].
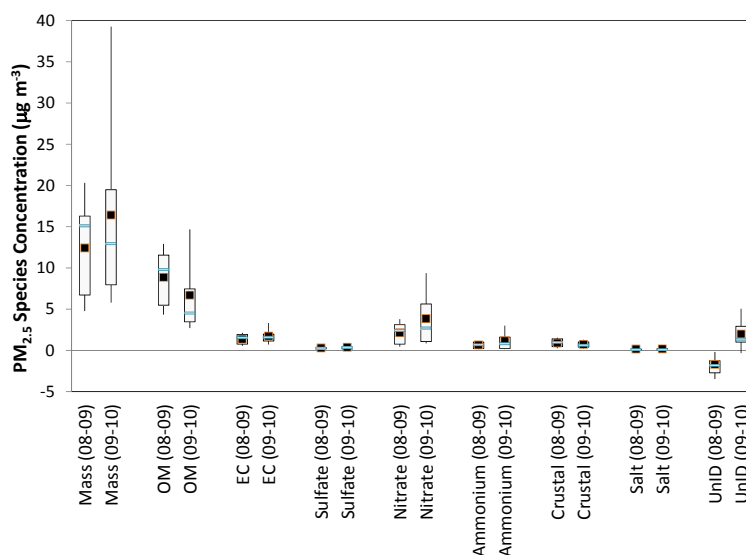

Fig. 5. Concentrations of PM$_{2.5}$ mass and major constituents (i.e., square: average; bar: median; box: 25th and 75th percentiles; whiskers: 10th and 90th percentiles) for winter 2008/2009 and 2009/2010. UnID is the unidentified mass (i.e., measured minus reconstructed PM$_{2.5}$ mass).

desorption-gas chromatography/mass spectrometry (TD-GC/MS; Ho and Yu, 2004; Ho et al., 2008, 2011; Chow et al., 2007d) for non-polar organic compounds, including ∼ 120 polycyclic aromatic hydrocarbons (PAHs), hopanes, sternanes, alkanes, and alkenes; (2) water extraction/total organic carbon (TOC; Yang et al., 2003) analysis for water-soluble organic carbon (WSOC); (3) ion chromatography-pulsed amperometric detection (IC-PAD; Engling et al., 2006) for levoglucosan; and (4) two-wavelength optical transmissometer (Ahmed et al., 2009; Chen et al., 2010a) for light absorption

(b_{abs}) at 370 and 880 nm. Twenty samples were analyzed for each of the species except for WSOC and levoglucosan on 18 December 2009 and 2 January 2010 where there were insufficient quartz-fiber filter remnants.

The temporal variations and relative abundances of organic species are presented in the Supplement, along with their two-month averages/signal-to-noise ratios (SNR; Fig. S1 and Table S1 of the Supplement). n-Alkanes are the most abundant organic compounds with concentrations ranging from 6.1 ng m^{-3} (20 January 2010)
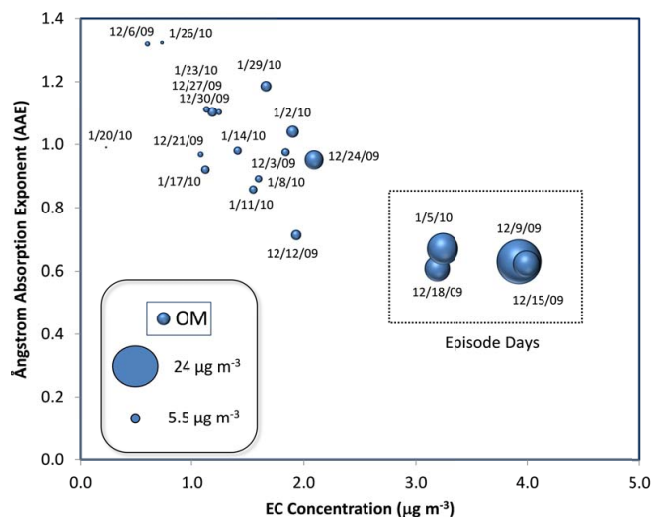
Fig. 6. Ångstrom Absorption Exponent (AAE) as a function of elemental carbon (EC) and organic material (OM) concentration. Diameter of the spheres is proportional to the OM concentration where $OM = 1.4 \times OC$. Episode days exceeding the USEPA 24-h $PM_{2.5}$ National Ambient Air Quality Standard (NAAQS) of $35\,\mu g\,m^{-3}$ are identified.

to $79.1\,ng\,m^{-3}$ (9 December 2009), containing abundant n-tetracosane (tetcos), n-hentriacontane (htricont), and n-pentacosane (pencos). The most abundant cyclo-/branched alkanes are anteiso-dotriacontane (anteisoc32), iso-tritriactotane (isoc33), and iso-hentriacotane (isoc33). Particle-bound PAHs (p-PAHs) are air toxics that derive from many combustion sources. The sum of measured p-PAH concentrations ranges from $0.82\,ng\,m^{-3}$ (2 January 2010) to $46.2\,ng\,m^{-3}$ (9 December 2009). Abundant p-PAH species include cyclopenta[c,d]pyrene (cp-cdpyr), retene, and benzo[b,j,k]fluoranthene (bbjkfl). Retene, a known marker for coniferous wood combustion (Ramdahl, 1983), is more enriched in the Truckee Meadows ($13 \pm 5\,\%$ of total p-PAHs) than in areas dominated by mobile sources (e.g., $1 \pm 1\,\%$ in El Paso, Texas; Chen et al., 2012a). Hopanes and steranes originate from oil combustion or from vehicle lube oil (Zielinska et al., 2008), and are $< 5\,ng\,m^{-3}$ with the dominant species being $\alpha\alpha\alpha$ 20S 24R/S-ethylcholestane (ster12), $17\alpha[H],21\beta[H]$-30-norhopane (hop17), and $17\alpha[H],21\beta[H]$-hopane (hop19).

Both WSOC and levoglucosan concentrations are high, particularly for the episode days. On 9 December 2009, WSOC reached almost $7\,\mu g\,m^{-3}$ while levoglucosan exceeded $1\,\mu g\,m^{-3}$. Levoglucosan has been considered as a primary marker for biomass burning (Fraser and Lakshmanan, 2000), and this supports the importance of residential wood combustion (RWC) contributions during the episode.

Aerosol b_{abs} at 370 and 880 nm correlate well with EC concentrations ($r^2 = 0.9$). The regression slopes are $21.3\,m^2\,g^{-1}$ for 370 nm and $13.6\,m^2\,g^{-1}$ for 880 nm. This is

consistent with EC as the dominant light-absorbing component. The Ångström absorption exponent (AAE) (Chen et al., 2006; Moosmüller et al., 2009):

$$AAE = \frac{\ln(b_{abs,370}) - \ln(b_{abs,880})}{\ln(370) - \ln(880)}, \qquad (4)$$

ranges from 0.61 to 1.32. Biomass burning aerosol from smoldering combustion generally shows AAE (> 1) higher than that from flaming combustion or motor vehicle exhausts with AAE < 1 (Kirchstetter et al., 2004; Chakrabarty et al., 2010; Chen et al., 2010a). In this study AAE appears to be lowest during the $PM_{2.5}$ episodes (Fig. 6), consistent with b_{abs} contributions mostly from flaming RWC and/or engine exhausts. The spectral characteristics of b_{abs} for the episode and non-episode periods are confirmed by in situ, continuous measurements at a nearby site (Gyawali et al., 2012). Smoldering contributions seem to be less important during the winter episodes.

3 Chemical mass balance source apportionment

The Effective Variance solution (Watson et al., 1984) to the CMB equations (EV-CMB) uses both source profiles and ambient concentrations as inputs to calculate source contribution estimates (SCEs) and associated uncertainties (Watson et al., 1984, 2008; Chow and Watson, 2002; Held et al., 2005; Chen et al., 2010b). EV-CMB was applied to each RENO_C speciated sample.

3.1 Source profiles

Source profiles relevant to sources in the Truckee Meadows are identified and referenced in supplemental Table S2. Major source types include: (1) geological materials; (2) motor vehicles (gasoline and diesel); (3) biomass burning (RWC and open burning); (4) cooking; (5) utility combustion; and (6) secondary aerosol, corresponding to the local $PM_{2.5}$ emission inventory (Ling-Barnes, 2010). Two fugitive dust samples (i.e., paved road dust – RNOPVRD, surface soils from a vacant lot – RNOSOIL) were obtained from representative locations around the RENO_C site along with two types of de-icing material (RNOSalt1 and RNOSalt2). These samples were dried, sieved, resuspended, and sampled onto filters (Chow et al., 1994) for chemical analyses identical to those for ambient samples, except that WSOC and levoglucosan were not quantified and set to zero owing to low OC contents (1.1–14.9 %, see Table S3).

Engine exhaust profiles considered include those acquired from the National Renewable Energy Laboratory Gas/Diesel Split Study (Fujita et al., 2007a, b) and the Las Vegas Carbon Source Apportionment Study (LVCSAS, Green et al., 2012b). These profiles are more recent and contain full organic speciation except for alkanes. They have been used in other EV-CMB applications (Chow et al., 2007c; Green et

al., 2012a). The Gas/Diesel Split profiles were based on dynamometer tests of 57 gasoline and 32 diesel vehicles while LVSAS adopted an in-plume monitoring system (Nussbaum et al., 2009; Zhu et al., 2009) at traffic intersections where air was dominated by a mixture of motor vehicle exhausts. Both studies were conducted in Nevada or neighboring California with vehicles and fuels similar to those of the Truckee Meadows.

The RWC source profiles resulted from tests during the Central California $PM_{10}/PM_{2.5}$ Air Quality Study (CRPAQS; Fitz et al., 2003) and the Lake Tahoe Source Characterization Study (LTSCS; Kuhns et al., 2004). CRPAQS focused on fireplace emissions while LTSCS tested both fireplaces and woodstoves. Both studies examined hardwood (i.e., oak, almond, and eucalyptus) and softwood (i.e., pine, tamarack, and juniper) combustion separately, resulting in six composite profiles. Levoglucosan's abundance ranges from 0.1 to 55.5 % among these six profiles. RWC emission compositions are highly variable depending on fuel and combustion conditions, as indicated by Chen et al. (2007a, 2010a). Open burning, including wildland fires, is not expected to contribute during winter months.

Due to the proximity of the RENO_C site to a Mexican restaurant, a meat cooking profile (BVCOOK, a composite of charbroiled chicken, chicken over propane, and charbroiled hamburger) as part of CRPAQS was included. Chow et al. (2007c) found meat cooking to be an important contributor at the Fresno supersite during winter. The markers for meat cooking are often associated with polar compounds such as cholesterol, palmitic acid, palmitoleic acid, stearic acid, and oleic acid (Fraser et al., 2003; Rinehart et al., 2006). However, the archived ambient samples were not amenable to analyses for these components.

Most of the reported point source profiles do not contain organic markers, but no large stationary emitters are located within the Truckee Meadows. Long-range transport from distant point sources is also low during winter. Point source profiles are therefore not included in the EV-CMB modeling. Secondary NO_3^- and $SO_4^=$ sources are represented by pure NH_4NO_3 (AMNIT) and $(NH_4)_2SO_4$ (AMSUL) profiles, respectively. Secondary OC is represented by a profile (SOC) containing exclusively (100 %) OC (Watson, 1979). This is equivalent to using the OC / EC ratio method to estimate SOC.

Table S1 indicates the EV-CMB fitting species. Important source markers that are commonly detectable in ambient samples and their corresponding sources include (e.g., Chow et al., 2007c; Watson et al., 2008): (1) Al, Si, Ca, Fe, Mn, K, Ti, and Sr for geological material/minerals; (2) EC and hopanes for diesel exhaust; (3) indeno[1,2,3,cd]pyrene (incdpy), benzo[g,h,i]perylene (bghipe), and coronene (corone) for gasoline engine exhaust; (4) K^+, retene, and levoglucosan for biomass burning; (5) pristane, and phytane for cooking; (6) Cu, Zn, As, Se, Br, Pb for industrial point sources; (7) Na^+, Cl for de-icing material; and (8) S, and NO_3^- for

secondary inorganics. OC is not a specific marker but it is a major $PM_{2.5}$ constituent. Temperature-resolved carbon fractions were not used as they were mostly determined by the IMPROVE (Chow et al., 1993) protocol in source profiles but by the IMPROVE_A (Chow et al., 2007a) protocol in ambient samples acquired after 2007. The two protocols are known to yield comparable OC and EC measurements but possibly differ for carbon fractions (Chow et al., 2005, 2007a; Chen et al., 2012b).

3.2 Sensitivity tests

EV-CMB sensitivity tests were applied to data from several samples to evaluate the performance of different source profile combinations in terms of correlation (r^2), root mean square difference (χ^2), and percent of mass explained (%MASS) (Watson and Chow, 2012). The initial source profile combination was modified in subsequent trials to examine changes in the SCEs and EV-CMB performance measures. Often, only one profile in each source type may be included since similar profiles result in collinearity, nonconvergence, and/or negative source contributions. An acceptable solution requires %MASS between 0.9 and 1.1, $r^2 > 0.9$, and $\chi^2 < 2$. The modified pseudo-inverse normalized (MPIN) matrix (Kim and Henry, 1999) would indicate the most influential species (e.g., MPIN value > 0.5) for each source type.

Table S4 illustrates two sensitivity test examples (an episode day on 5 January 2010 and a median loading day on 17 January 2010). Better EV-CMB fits to the data result from a combination of paved road dust (RNOPVRD), road salt (RNOSALT2), mobile source (gasoline and diesel engine exhaust; GAS and DIESEL, respectively), RWC (fireplace and woodstove burning hardwoods; CRBURN_H and LTWS_H), and secondary (AMNIT and AMSUL) source profiles. The DIESEL profile may represent both on-road and off-road diesel engine emissions. Gasoline-engine exhaust contributions appear to be minor compared with those from diesel engines. Including softwood RWC profiles in the CMB model always lowers the overall model performance.

Including the cooking (BVCOOK) profile improves performance measures (e.g., lower χ^2 and higher %MASS). However, the cooking contribution would be comparable to or even greater than the combined contribution from gasoline and diesel engine exhaust (Table S4). The MPIN matrix indicates that OC is the only species influencing the cooking source. Due to the lack of more specific cooking markers such as cholesterol and palmitoleic acid, BVCOOK was deselected. Adding SOC into the mix led to non-convergence for both samples.

The MPIN matrices for the selected source combination (Table S5) indicates that the most important diesel engine exhaust marker is EC, followed by Zn while the gasoline engine exhaust contribution is marked by coronene. indeno[1,2,3,cd]pyrene and benzo[g,h,i]perylene are more

associated with the wood stove profile (i.e., LTWS_H), along with levoglucosan, OC, and $17\alpha(H),21\beta(H)$-hopane. This explains the low gasoline engine exhaust SCEs by EV-CMB. The fireplace emission (i.e., CRBURN_H) is marked by retene and K^+. Si is the most influential species for paved road dust profile (i.e., RNOPVRD), followed by Al and Ca. Fe, however, is associated with a de-icing profile (i.e., RNOSalt2) along with Na^+ and Cl. Water-soluble iron cyanide compounds are widely used as anticaking agents in road salt. NO_3^- and S are unambiguous markers for secondary NH_4NO_3 and $(NH_4)_2SO_4$, respectively.

Figure 7 compares measured and calculated $PM_{2.5}$ species concentrations. For the 5 January 2010 sample, the EV-CMB solution explains measured species well except for the carbon fractions (EC1, EC2, and OP, not included in the fitting but calculated from SCEs), Pb, As, $22R-17\alpha(H),21\beta(H)$-30,31,32-Trishomohopane (hop27), and phytane. In addition, Cl, Ti, Br, Cu, Rb, and all hopanes were not fitted as well for the 17 January 2010 $PM_{2.5}$ sample. None of these species, however, are unique source markers. Removing these species somewhat alters the SCEs but the changes are within the uncertainty intervals.

3.3   Source contribution estimates

Figure 8 shows the winter 2009/2010 SCEs for $PM_{2.5}$, OC, and EC. RWC is resolved for all samples, though the partition between fireplace and woodstove contributions varies. Woodstove contributions are missing for the 18 December 2009 and 2 January 2010 samples because levoglucosan data were not available on these two days. Diesel contributions show up every day except for 26 January 2010 when only gasoline engine exhaust was detected. This abnormality is hard to explain physically, and it is a reminder of the uncertainty inherent in separating diesel and gasoline source contributions even with organic markers. De-icing contributions are also resolved for every sample except 26 January 2010, while paved road dust does not appear on 3–9, 15–18, and 30 December 2009. De-icing material contributed more to $PM_{2.5}$ than paved road dust during December 2009, while the opposite was true during January 2010. This is consistent with more snowfalls in December 2009 since snow covers reduce fugitive dust but increase salting and de-icing activities. Secondary $(NH_4)_2SO_4$ is resolved for all samples except for 2 January 2010, although its contribution never exceeds $0.25\,\mu g\,m^{-3}$. EV-CMB-calculated $PM_{2.5}$ mass concentrations are generally lower than measured $PM_{2.5}$. For the two days with poor mass closure (30 December 2009 and 20 January 2010), model calculated $PM_{2.5}$ mass are closer to the reconstructed mass than the gravimetric mass. A strong correlation ($r^2 = 0.89$) between the unexplained $PM_{2.5}$ mass and secondary NH_4NO_3 (AMNIT) contribution supports water associated with NH_4NO_3 as a major component of unexplained mass (Fig. 9).

RWC, diesel engine exhaust, and secondary NH_4NO_3 were the major $PM_{2.5}$ contributors during NAAQS exceedances. The highest RWC contribution of $26.6\,\mu g\,m^{-3}$ or 51 % of $PM_{2.5}$ mass occurred on 9 December 2009. Most of the RWC was attributed to fireplace emissions. An intense and persistent inversion layer and deep snow cover, coupled with low temperatures during the day (daily mean: $-13\,°C$), enhanced RWC emissions and accumulation. Secondary NH_4NO_3 accounted for 26.7 % of $PM_{2.5}$ mass on 9 December 2009. Particulate NH_4NO_3 is also favored by low temperatures and high RH (Watson et al., 1994; West et al., 1999; Chow et al., 2008). Diesel engine exhaust explained another 6.5 % of $PM_{2.5}$ while other sources were minor contributors (0–3 %). For the other episode days (15, 18 December 2009, and 5 January 2010), RWC contributions were lower (11–39 %) while diesel contributions were higher (13–22 %). Higher ambient temperatures, i.e., daily mean: 0–1 °C, compared to those of 9 December 2009 may explain the difference. Nevertheless, ambient temperatures on the three days appear to be low enough for NH_4NO_3 formation – NH_4NO_3 fractions ranged from 27 to 37 %. Paved road dust was an important contributor (5.7 %) only on 5 January 2010.

RWC from fireplaces also dominated the ambient OC concentrations (Fig. 8). On 15 and 18 December 2009, diesel exhaust was almost as important as RWC as an OC source. The unexplained OC, up to 44 % of measured OC, may be in part attributed to a positive organic sampling artifact, which has not been adequately corrected on the STN samples (Chow et al., 2010). Since OC was quantified on quartz-fiber filters and mass was measured on Teflon-membrane filters, this artifact should not contribute to the unexplained $PM_{2.5}$ mass. Certainly the missing OC might also result from SOC and/or differences between the selected source profiles and the actual (but unknown) profiles typical of local RWC emissions. The EV-CMB model explains EC better than OC. There is no organic sampling artifact associated with EC. Diesel engine exhaust is the dominant EC source.

3.4   Origins of secondary ammonium nitrate

Comparing $PM_{2.5}$ chemical speciation of winter 2009/2010 with winter 2008/2009 shows that the higher secondary NH_4NO_3 lifted $PM_{2.5}$ concentrations over the $35\,\mu g\,m^{-3}$ threshold. The formation of NH_4NO_3 may depend on gaseous ammonia (NH_3) or nitric acid (HNO_3; derived from NO_x emissions) concentrations and be influenced by ambient temperature and RH (Watson and Chow, 2002). An ion balance shows a full neutralization of NO_3^-, and SO_4^{2-} by NH_4^+ during the $PM_{2.5}$ episodes and so NO_x emissions and oxidation likely limit NH_4NO_3 formation in the valley. A good correlation of 24-h NO_x and AMNIT ($r^2 = 0.82$) is found for winter 2009/2010.

Most wintertime NO_x is generated from local sources, particularly mobile and RWC emissions, and so is NO_3^-.
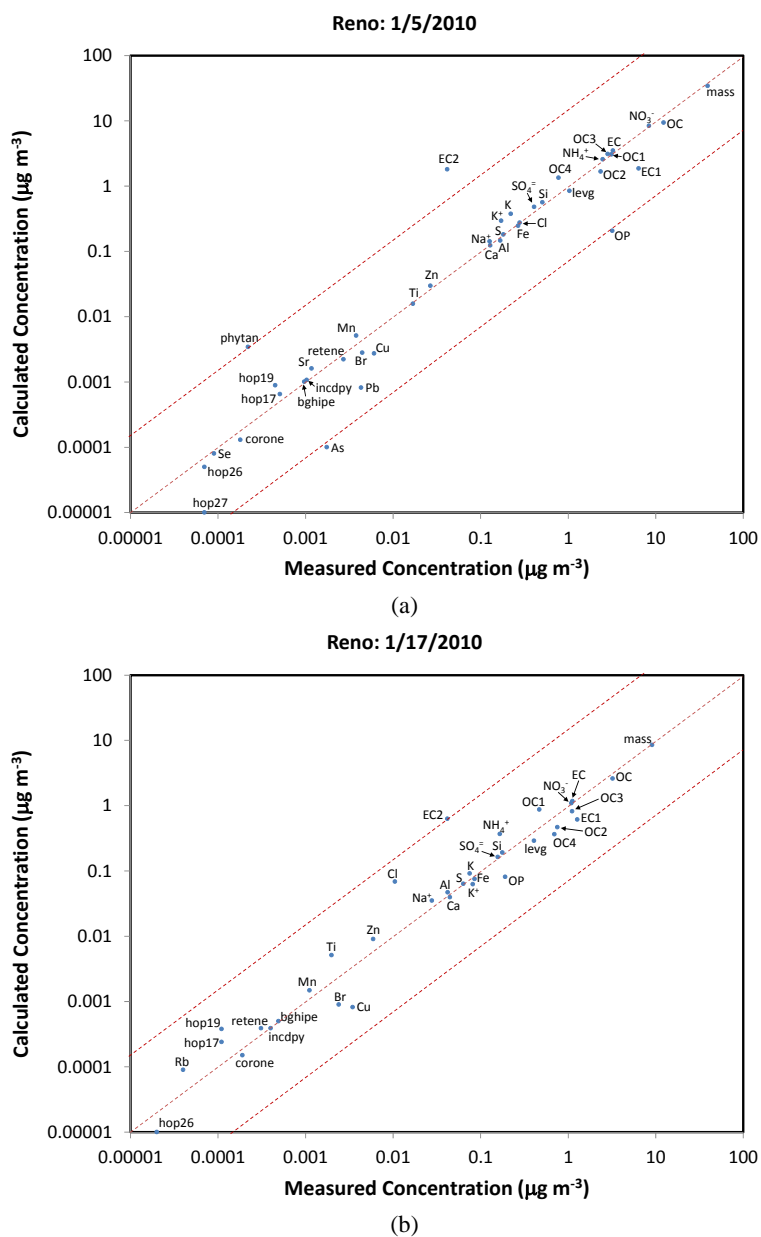
Fig. 7. Comparisons of EV-CMB calculated versus measured PM$_{2.5}$ species for: **(a)** 5 January 2010 and **(b)** 17 January 2010 samples from the Reno monitoring site (RENO_C). See Supplement Table S0 for definitions of organic species abbreviations.

Compression-ignition engines used in diesel vehicles have long been considered important NO$_x$ sources owing to a high combustion temperature (Lloyd and Cackette, 2001; Chow, 2001). Biomass burning, especially high-temperature flaming combustion, may also generate substantial NO$_x$ (Chen et al., 2007a, 2010a). Other Truckee Meadows NO$_x$ sources include natural gas combustion in furnaces and boilers (Ling-Barnes, 2010), which have higher emissions during winter than other seasons. Coal-fired power plant contributions and other distant sources can be excluded as NO$_x$ and NO$_3^-$ con-

tributors during winter owing to their great distance from the Truckee Meadows.

To a first approximation, secondary NH$_4$NO$_3$ affiliated with mobile sources or RWC was estimated from regressing AMNIT contributions against mobile and RWC contributions. A robust regression method (Huber, 1981) was applied which involves iterative reweighting of the input data values to lessen the influence of extreme values. The two RWC sources (i.e., CRBURN_H and LTWS_H) were combined to form an overall RWC contribution while the diesel and gasoline sources (i.e., DIESEL and GAS) were pooled to
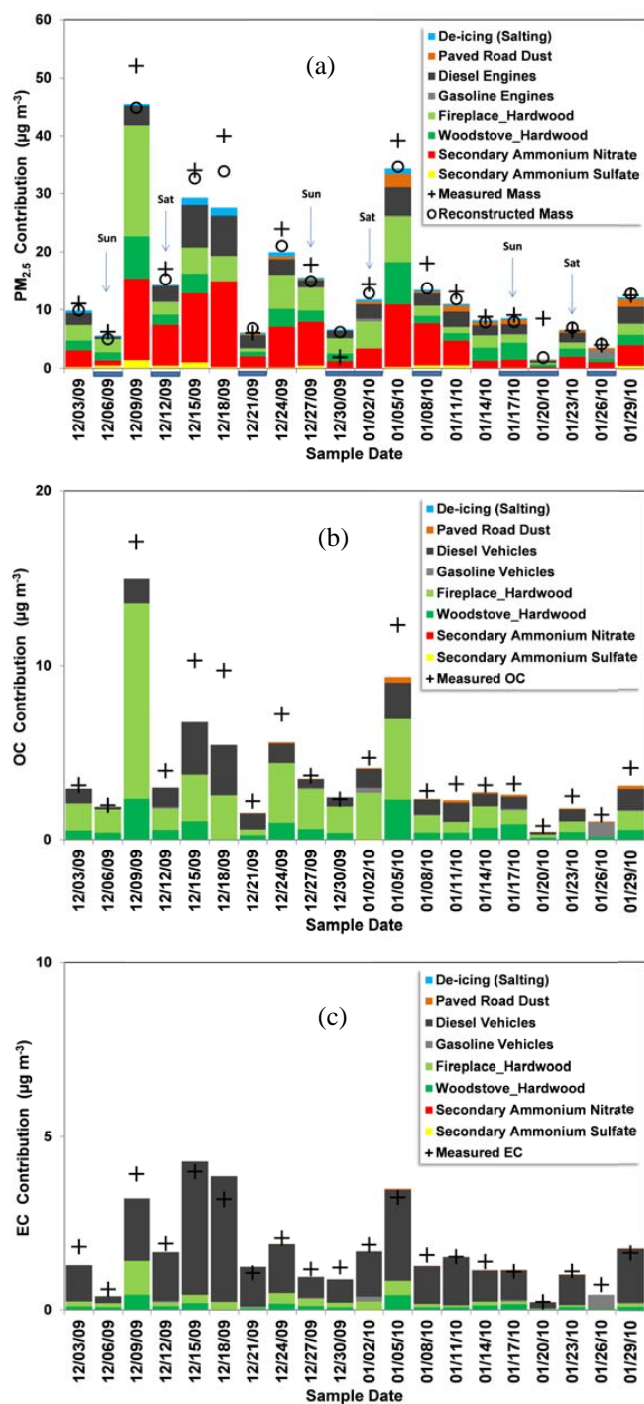
Fig. 8. EV-CMB PM$_{2.5}$ source apportionment for: **(a)** mass, **(b)** OC, and **(c)** EC. Stacked columns indicate model estimates, which are compared with measured and reconstructedmass concentrations. Weekends are noted in **(a)**. Snowy days are marked by a blue bar under the column in **(a)**. Snow cover typically accumulates during or after the snowfall.
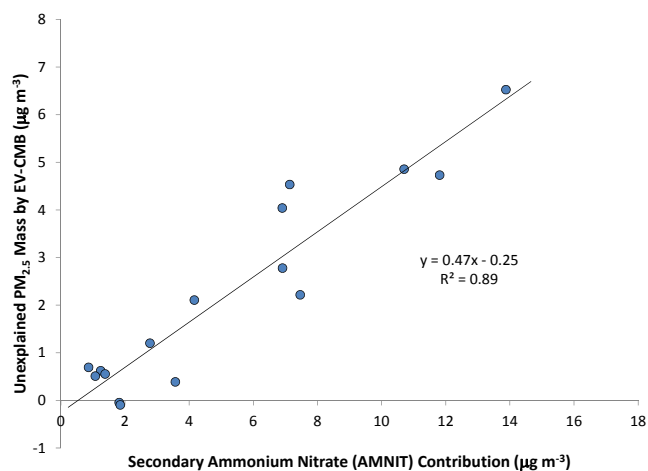

Fig. 9. Relationship between unexplained PM$_{2.5}$ mass and secondary ammonium nitrate contribution by EV-CMB analysis. Four data points were excluded due to either missing levoglucosan measurements (18 December 2009 and 2 January 2009) or very poor mass closure (30 December 2009 and 20 January 2010).
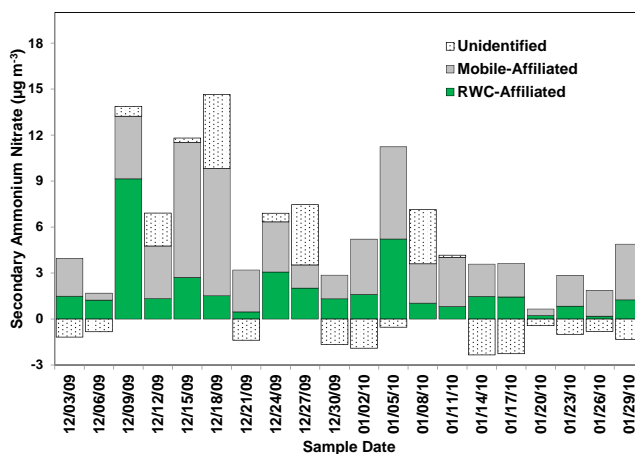

Fig. 10. Source apportionment of secondary ammonium nitrate by robust multiple linear regression.

represent the mobile contribution. The regression of AMNIT against the RWC and mobile contributions together yields a good correlation ($r^2 = 0.81$), better than the regression correlations against RWC or mobile contributions alone ($r^2 = 0.45$–0.65). The regression coefficients are 0.34 and 1.20 for RWC and mobile sources, respectively, suggesting that 0.34 and 1.2 µg m^{-3} secondary NH$_4$NO$_3$ is produced per µg m^{-3} of primary PM$_{2.5}$ emitted from the RWC and mobile sources, respectively.

Figure 10 shows mobile- and RWC-affiliated secondary NH$_4$NO$_3$, which explain total NH$_4$NO$_3$ (i.e., AMNIT) levels for three of four high PM$_{2.5}$ days: 9, 15 December 2009, and 5 January 2010. (Note: the RWC contribution on 18 December 2009 is underestimated due to missing levoglucosan

measurement.) Although the mobile sources did not contribute to primary $PM_{2.5}$ as much as RWC during winter 2009/2010, the mobile-source contribution might be responsible for $\sim 2/3$ of secondary NH_4NO_3.

4   Conclusions

The Truckee Meadows is an urban valley in the arid west of the US with light-to-moderate snowfall and frequent subsidence in winter. The formation of $PM_{2.5}$ episodes shows meteorological and chemical characteristics that are likely common for many urban valleys in the Western Cordillera.

First, the highest $PM_{2.5}$ concentrations are accompanied by intense and prolonged (multi-day) temperature inversions, snow on the ground, and low wind speeds. These variables are inter-related, as snow cover increases the reflectance of sunlight, thereby decreasing heating at the surface that would break up the inversion. Most of the surface heating that remains is absorbed as latent heat, melting the snow rather than raising the surface temperature. The prolonged inversion creates a stagnant condition that inhibits ventilation by wind.

Second, OC and EC dominate the $PM_{2.5}$ mass, but during episodes much elevated NH_4NO_3 and unidentified mass concentrations drive $PM_{2.5}$ levels over the $35\,\mu g\,m^{-3}$ threshold. The low temperatures and higher RH associated with snow cover and melting snow encourage a shift in equilibrium for gaseous HNO_3 and NH_3 to form particulate NH_4NO_3, thereby increasing its contribution to $PM_{2.5}$ mass. Liquid water associated with NH_4NO_3 explains most unidentified mass in the samples. The water may not completely leave these particles until RH decreases below 30 %.

An EV-CMB receptor model suggests that RWC, diesel exhaust, and NH_4NO_3 were major contributors to $PM_{2.5}$ in the Truckee Meadows during winter 2009/2010. RWC contributions ranged from 11–51 % of $PM_{2.5}$ mass on four $35\,\mu g\,m^{-3}$ exceedance days (9, 15, 18 December 2009, and 5 January 2010). Most of the RWC was attributed to fireplaces burning hardwoods. The wide range of RWC contribution may reflect different ambient temperatures, as lower temperatures increase home heating demand. Secondary NH_4NO_3 accounted for 27–37 % of $PM_{2.5}$ while diesel engine exhaust explained 7–22 % of $PM_{2.5}$. Paved road dust and de-icing materials were minor, but detectable, contributors. Gasoline engine exhaust was a small contributor, as was secondary $(NH_4)_2SO_4$ that never exceeded a $0.25\,\mu g\,m^{-3}$ contribution to $PM_{2.5}$ mass. Contributions from large industrial sources (e.g., coal-fired power plants) were not quantified, though all the known large point sources are distant from the Truckee Meadows and transport is low during winter stagnations. Neither cooking nor SOC contributions were resolved.

A multiple linear regression of the secondary NH_4NO_3 contribution on the mobile source and wood burning contributions suggests that about 2/3 of the NH_4NO_3 is associated with mobile source emissions and 1/3 with RWC emissions,

consistent with most of the NO_3^- deriving from engine NO_x emissions. Reducing use of diesel-powered equipment and restricting domestic burning for unfavorable pollution meteorology forecasts should contribute to the prevention of future NAAQS exceedances.

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