

Appendix G

Chemical Analysis of PM10 and PM2.5 Filters from Mammoth Lakes (DRI, 2013)

**Chemical Analysis of PM10 and PM2.5 filters from
Mammoth Lakes**

Final Report

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Prepared for the Town of Mammoth Lakes

May 21, 2013

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Introduction

A study was conducted to try to gain understanding regarding source contributions to high PM10 values recorded at the Mammoth Lakes Gateway air quality monitoring site.

DRI proposed to analyze fifteen PM10 and five PM2.5 filters (dates and concentrations shown in Tables 1 and 2) for elements and ions. The fifteen PM10 filters included the nine highest and 11th highest PM10 filter concentration days since 2001, eight of which were since the start of 2008. The other five PM10 filters are days for which high PM10 and/or PM2.5 concentrations were measured and both PM10 and PM2.5 filters are available for each day. Three of these five days had the highest PM10 for days with PM2.5 filters also. The other two days had high PM2.5 and high PM2.5 to PM10 ratios (indicative of large relative wood smoke impact). Because the road dust is expected to be mostly in the larger sizes and wood smoke is mostly in the small sizes (PM2.5) analyzing both PM10 and PM2.5 for a few days will be informative and will provide supporting evidence for our estimates based on the PM10 data.

The samples proposed to be analyzed are listed below.

Table 1. Dates with only PM10 filters:

Date	PM10 concentration
12/21/2001	134.2
2/13/2002	129.3
1/1/2008	85.9
2/9/2008	96.7
2/27/2008	95.5
2/21/2009	117.6
1/2/2010	101.3
1/29/2010	104.0
2/13/2010	92.0
1/12/2011	127.6

Table 2. Dates with PM10 and PM2.5 filters:

Date	PM10	PM2.5
3/14/2001	55.0	35.2
12/17/2003	74.6	33.5
1/13/2005	39.3	27.0
1/19/2005	85.1	25.2
1/22/2005	77.8	27.4

Filters for the three days in 2001 and 2002 were not provided to DRI for analysis. Chemical analysis of filters was done on the remaining samples listed above, and included twelve PM10 filters and four PM2.5 filters.

The chemical analysis included X-ray fluorescence (XRF, giving most elements), ion chromatography (anion and cations), and light absorption. The XRF analysis provides the road dust elements, ion chromatography the contributions from sulfate and nitrate, and ammonium (associated with nitrate and sulfate). The light absorption measurement is a good indicator of elemental carbon (EC) (diesel exhaust and wood smoke primarily). What the analysis cannot do that is important is organic carbon (OC), because the Teflon filters that have been collected cannot provide that. Organic carbon was estimated as a residual.

Chemical analysis results

Total PM₁₀ concentrations and component concentrations by date are shown in Table 3.

Table 3. PM10 concentrations and measured or reconstructed major species concentrations by date. The residual is calculated as the total PM10 concentration – (soil+SO₄+NO₃+NH₄+sea salt+ EC). It was assumed that all unaccounted for mass is organic mass (OM) and the ratio of organic mass to organic carbon is 1.8.

Date	conc stp	soil	SO4	NO3	NH4	seasalt	EC	residual	OC
12/17/03	74.6	22.3	0.32	0.64	0.02	0.30	9.1	41.9	23.3
01/13/05	39.3	5.5	0.28	0.40	0.02	0.07	10.2	22.8	12.7
01/19/05	85.1	34.1	0.33	0.49	0.02	0.25	9.0	40.9	22.7
01/22/05	77.8	28.9	0.27	0.40	0.00	0.28	8.3	39.7	22.0
01/01/08	85.9	44.3	0.60	0.69	0.03	0.37	9.8	30.2	16.8
02/09/08	96.7	25.2	0.30	0.38	0.02	0.20	6.8	63.8	35.4
02/27/08	95.5	44.6	0.40	0.38	0.05	0.11	7.5	42.5	23.6
02/21/09	117.6	51.6	0.79	0.80	0.05	1.12	7.6	55.6	30.9
01/02/10	101.3	44.6	0.29	0.48	0.00	2.68	7.6	45.6	25.4
01/29/10	104.0	47.3	0.36	0.54	0.04	0.68	6.7	48.4	26.9
02/13/10	92.0	41.2	1.10	0.50	0.23	0.34	7.9	40.7	22.6
01/12/11	127.6	4.3	0.16	0.16	0.00	0.07	5.4	117.4	65.2

Because only Teflon filters were available, OC and EC could not be analyzed. Reconstructed mass calculations (using the IMPROVE protocol, Pitchford et al., 2007) showed that for most days, reconstructed mass was far less than measured mass. This is expected when there is a significant contribution from carbonaceous aerosol as with residential wood combustion, and other ambient sources of carbon, such as in road dust.

Elemental carbon was estimated from filter light absorption (Chow et al., 2010) as $EC = (b_{abs})/10$ where b_{abs} is filter light absorption in inverse megameters (Mm^{-1}) and EC concentration is in $\mu g/m^3$. EC concentration was then added to the reconstructed mass. The residual of measured

mass – reconstructed mass was assumed to be organic mass (OM), which was assumed to be 1.8 times OC (as used in the IMPROVE algorithm). Thus OC is the residual/1.8.

From Table 3 it can be readily seen that OC, EC, and soil are the only significant contributors to PM10 concentrations. The nitrate is expected to be biased low as it tends to volatilize off the filter, especially after exposure to x-rays in a vacuum as occurs during the XRF analysis prior to analyzing for nitrate with ion chromatography. It should be noted here that the PM10 concentration of 127.6 $\mu\text{g}/\text{m}^3$ on 1/12/2011 is probably in error. The soil and EC concentrations are not consistent with such a high mass concentration and the TEOM FDMS data for the day was only 27.7 $\mu\text{g}/\text{m}^3$. The TEOM FDMS and filter PM10 were usually close in magnitude.

Table 4 shows the same information as Table 3 for the PM_{2.5} samples.

Table 4. PM_{2.5} concentrations and measured or reconstructed major species concentrations by date. To be comparable to the PM10 data, the concentrations have been adjusted to standard temperature and pressure as defined by EPA (25°C, 760mm Hg).

Date	conc stp	soil	SO4	NO3	NH4	seasalt	EC	residual	OC
12/17/03	40.6	1.19	0.24	0.45	0.03	0.12	7.8	30.8	17.1
1/13/05	32.7	0.50	0.24	0.39	0.03	0.04	10.2	21.4	11.9
1/19/05	30.5	1.89	0.26	0.28	0.03	0.07	8.8	19.2	10.7
1/22/05	33.2	1.60	0.21	0.29	0.02	0.08	8.3	22.7	12.6

For PM_{2.5}, most of the mass is EC and residual (assumed OM). Table 5 shows the percentage of each chemical component that was in the fine mode (PM_{2.5}) for the four days with both PM₁₀ and PM_{2.5} chemically speciated data.

Table 5. Percent of each chemical component that is in the fine mode.

	PM10 stp	soil	SO4	NO3	NH4	seasalt	EC	OC
12/17/03	54.4	5.3	76.4	70.3	144.8	40.1	85.1	73.4
1/13/05	83.2	9.0	86.9	97.8	158.6	60.0	99.8	93.5
1/19/05	35.9	5.5	78.1	57.5	133.1	29.3	97.9	46.9
1/22/05	42.7	5.5	75.1	70.8		27.4	100.7	57.1

As expected most of the sulfate, nitrate, and EC are in the fine mode and most of the soil is in the coarse mode. For 12/17/03 and 1/13/05 most of the OC is in the fine mode as would be expected with wood smoke. For 1/19/05 and 1/22/05 only about half to a little more than half of the estimated OC is in the fine mode. It should be noted that the fine/coarse mode OC split is highly uncertain because OC in both size ranges was estimated from a residual.

Diurnal patterns in PM10 concentrations

Consideration of diurnal patterns in PM₁₀ concentrations may provide insight into likely sources of high PM10. Hourly PM10 concentrations for chemical analysis days in 2009 and 2010 are

shown in Figure 1. This period had hourly measurements using the TEOM FDMS monitor which gives values similar to the filter based concentration measurements (Table 6). All days had morning and evening peaks although the timing of the peaks varied.

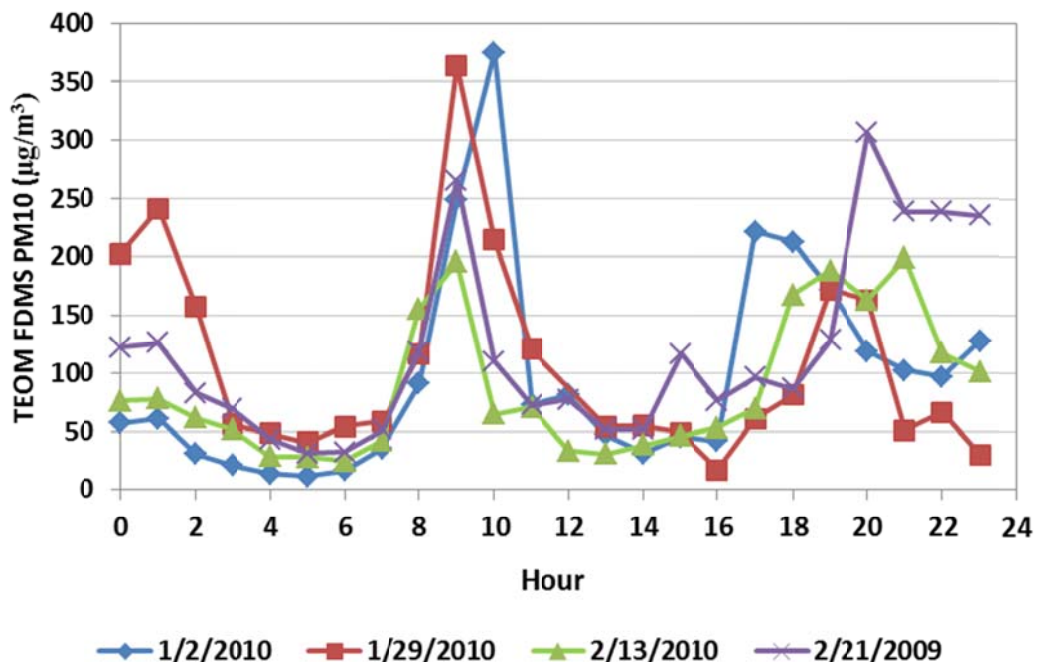


Figure 1. TEOM FDMS hourly concentrations for 1/2/2010, 1/29/2010, 2/13/2010 and 2/21/2009.

Table 6. Comparison of filter PM10 and TEOM PM10 for days with chemical speciation. For days before 2009 a standard TEOM was used. From 2009 and after, a TEOM FDMS was used.

Date	Filter	TEOM	TEOM/Filter
12/17/2003	74.6	60.4	0.81
1/13/2005	39.3	29.2	0.74
1/19/2005	85.1	46	0.54
1/22/2005	77.8	40.1	0.52
2/21/2009	117.6	116.2	0.99
1/2/2010	101.3	97.3	0.96
1/29/2010	104	107.6	1.03
2/13/2010	92	87	0.95
1/12/2011	127.6	26.8	0.21

Figure 2 gives hourly PM10 concentrations for days in 2003 and 2005 with chemical speciation measurements. These were measured using a standard TEOM which heats the sample to 50°C which drives off most of the volatile compounds including much of the organic material in wood

smoke and ammonium nitrate. During these days, the TEOM PM10 was much less than the filter-based PM10 as shown in Table 6.

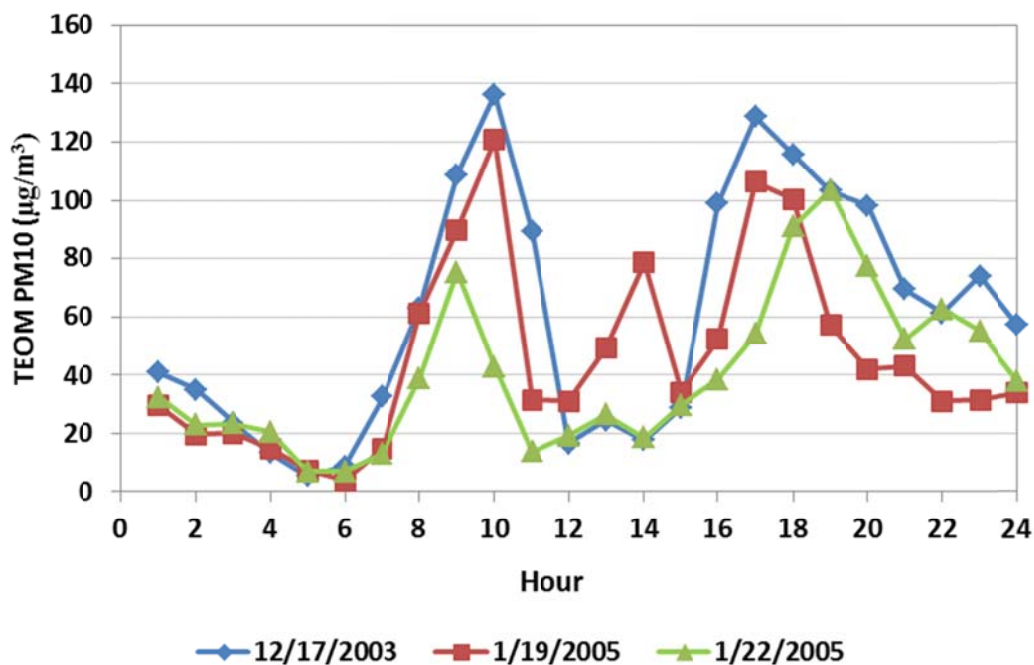


Figure 2. TEOM hourly PM10 concentrations for 12/17/2003, 1/19/2005, and 1/22/2005.

Figure 3 shows average TEOM PM10 by hour of day for winter months (December, January, and February) for the 2001-2008 and 2009-2012 periods. This essentially corresponds to the time frame with standard TEOM (2001-2008) and the period with TEOM FDMS (2009-2012). The concentrations are similar and diurnal patterns both show morning and evening peaks. The latter period shows somewhat higher concentrations overnight (from about 9pm to 8 am). This is expected because the standard TEOM volatilizes a portion of the wood smoke. The morning peaks are of very similar concentration both periods, suggesting that this peak is almost entirely traffic related (e.g. road dust) and that concentrations have changed little over time. It is possible however that volatile compounds (e.g. wood smoke) also contribute to the morning peak and the levels have decreased by an amount equal to the volatile component of the wood smoke lost by the standard TEOM, thus masking any trend in time. We can also note that the earlier evening peak in PM10 seen gives the same concentrations for both time periods until after 9pm when the 2009-2012 period shows higher values likely due to less volatilization of wood smoke from the sample. This suggests that the influence of wood smoke relative to road dust increases during late evening to morning and that the earlier rise in PM10 may be driven mainly by road dust.

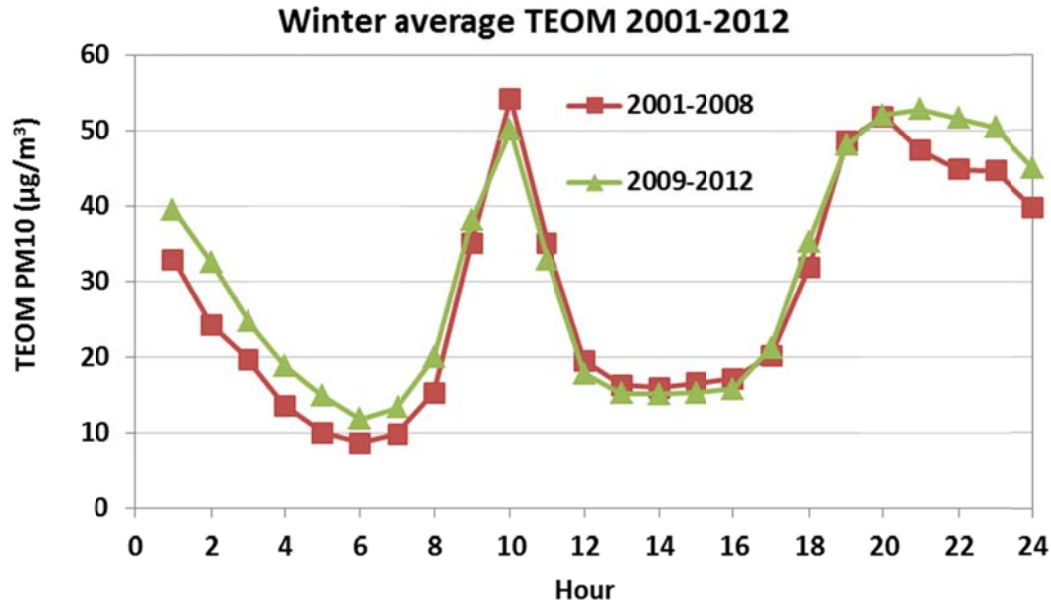


Figure 3. Average PM₁₀ by hour of day for winter months (December, January, and February) for the 2001-2008 and 2009-2012 periods. The 2009-2012 period had TEOM FDMS; the 2001-2008 period mostly was standard TEOM.

Methodology for CMB 8 runs for Mammoth Lakes PM10 and PM2.5

Chemical mass balance (CMB) version 8 (USEPA) was used to estimate source contributions to PM₁₀ and PM_{2.5} for days with chemically speciated data. CMB analysis done in support of the 1990 Air Quality Management Plan (AQMP, Ono et al, 1990) for Mammoth Lakes showed most of the PM₁₀ was due to road dust and residential wood combustion (RWC). One of the goals of the current study is to try to determine if the relative contribution of RWC and road dust has changed since the 1990 report.

Source profiles used for CMB

PM10 analysis

Source profiles were taken from the 1990 AQMP. Profiles for Mammoth Lakes woodstoves and fireplaces were used plus road dust and cinders. When running CMB it was found that significant collinearity occurred between the wood stove and fireplace sources, giving high uncertainty to wood stove contributions (sometimes significantly negative). CMB was run again not using the wood stove source, just fireplaces, cinders, and road dust. This gave much improved results. It was attempted to improve results by averaging the wood stove and fireplace profiles, but the quality of the results deteriorated. A weighted average of 3 parts fireplace to 1 part wood stove profile was also tried and results were not as good as simply using the fireplace profile.

PM2.5 analysis

The PM2.5 analysis had collinearity problems between wood stove and fireplace and between road dust and cinders. The analysis with the best results used fireplaces and road dust source profiles only.

Fitting species

CMB results can vary significantly based on the choice of fitting species selected. In the initial run, all species measured with available source analysis were used. This gave poor results. Species that were noted from the ambient data to represent the mix of sources were added and subtracted in a trial and error method until the best results were obtained. Attention was paid to squared correlation coefficient, the Chi square statistic and percent of mass explained performance measures (Watson, 2004). The fitting species included: soluble Cl, ammonium, soluble K, total K, Na, Al, Si, S, Ca, Ti, Fe, EC, OC, Ba, Sr, and Zn.

CMB results

Results for the best fitting CMB analysis are shown in Table 7. Recommended performance measures (Watson, 2004) are % mass accounted for $100\pm 20\%$, $R\text{-squared} > 0.80$, and $\text{Chi-squared} < 4.0$. For the PM10 results (discounting the 1/12/2010 sample), 10/11 cases have % mass $100\pm 20\%$, all cases have $R\text{-squared} > 0.80$, and 5/11 meet the $\text{Chi-squared} < 4.0$ criteria. For the PM2.5 results, three out of four met the mass criteria and all four met the $R\text{-squared}$ and chi-squared criteria.

Table 7. CMB attribution results and performance statistics.

DATE	SIZE	conc stp	FP	RD	CIND	SUM	% mass	R ²	Chi ²
12/17/03	10	74.6	53.0	9.6	10.2	72.8	97.5	0.94	3.70
01/13/05	10	39.3	31.1	3.6	1.7	36.5	92.7	0.92	3.76
01/19/05	10	85.1	51.8	18.1	11.5	81.3	95.5	0.93	4.39
01/22/05	10	77.8	47.5	16.9	8.7	73.0	93.8	0.94	3.70
01/01/08	10	85.9	53.4	24.6	14.8	92.9	108.1	0.93	4.73
02/09/08	10	96.7	68.0	10.2	13.8	92.0	95.1	0.94	3.35
02/27/08	10	95.5	52.6	26.1	12.7	91.3	95.6	0.90	6.40
02/21/09	10	117.6	91.7	26.6	21.0	139.3	118.5	0.94	3.89
01/02/10	10	101.3	100.4	23.2	19.0	142.6	140.8	0.90	6.88
01/29/10	10	104.0	65.8	24.5	16.8	107.1	103.0	0.93	4.52
02/13/10	10	92.0	68.9	25.9	13.9	108.7	118.2	0.91	6.09
01/12/11	10	127.6	43.9	2.2	1.8	47.9	37.5	0.82	8.77
12/17/03	2.5	40.6	28.6	1.5		30.0	74.0	0.96	0.90
01/13/05	2.5	32.7	27.5	0.6		28.1	86.0	0.81	3.90
01/19/05	2.5	30.5	28.1	2.2		30.3	99.3	0.89	2.73
01/22/05	2.5	33.2	27.6	1.9		29.5	89.0	0.92	2.00

The percent of each sample attributed to each source is shown in table 8. On average, residential wood combustion contributed about 2/3 of the PM10, while traffic (road dust + cinders) contributed about 1/3, according to the CMB results.

Table 8. Percent of each sample attributed to fireplaces, road dust, and cinders. January 12, 2011 not included as this date had an apparent mass problem as discussed earlier. January 13, 2005 was not included in the PM10 list because it had low PM10 concentrations and thus does not represent a high PM10 day.

DATE	SIZE	conc stp	FP%	RD%	CIND%
12/17/03	10	74.6	72.9	13.2	14.0
01/19/05	10	85.1	85.3	10.0	4.7
01/22/05	10	77.8	63.7	22.2	14.1
01/01/08	10	85.9	65.0	23.1	11.8
02/09/08	10	96.7	57.5	26.5	16.0
02/27/08	10	95.5	73.9	11.1	15.0
02/21/09	10	117.6	57.5	28.5	13.9
01/02/10	10	101.3	65.8	19.1	15.1
01/29/10	10	104.0	70.4	16.3	13.3
02/13/10	10	92.0	61.4	22.9	15.7
12/17/03	2.5	40.6	28.6	4.5	
01/13/05	2.5	32.7	95.2	4.8	
01/19/05	2.5	30.5	97.7	2.3	
01/22/05	2.5	33.2	92.6	7.4	
Average PM10		93.1	67.4	19.3	13.4
Average PM2.5		34.3	78.5	4.7	

For the winter 1987-1988 study (Ono et al, 1990), on average, fireplaces contributed 75% of the PM10 and road dust 25%. Table 9 compares the results of the 1987-88 study to this study. Three of the days in the 1987-88 study showed wood smoke contributing >95% of the PM10; the current study shows no high PM10 days with greater than 75% of the PM10 contributed from wood smoke.

Furthermore, the peak contributions of road dust and residential wood combustion to PM10 appear to have diminished. For the 1987-88 study average contributions to PM10 from road dust and fireplaces for the three highest impact days were 72 and 107 $\mu\text{g}/\text{m}^3$, respectively. For the 2008-2010 period these fell to 56 and 72 $\mu\text{g}/\text{m}^3$. This represents a 32% drop in road dust concentrations and 33% drop in residential wood combustion concentrations on high impact days.

Table 9. CMB percent contributions to PM10 by road dust (RD) and residential wood combustion (RWC) for the winter 1987-88 study and this study (2003-2011). Road dust and cinders contributions are combined into RD .

Date	Conc	RD%	RWC%
12/26/1987	125.9	2.3	97.7
12/30/1987	132.8	1.3	98.7
12/31/1987	142.8	2.5	97.5
1/1/1988	117.4	10.3	89.7
1/22/1988	143.8	33.7	66.3
1/23/1988	157.8	41.2	58.8
2/3/1988	104.3	31.5	68.5
2/5/1988	148.2	33.8	66.2
2/6/1988	160	31.2	68.8
2/13/1988	137.6	38.8	61.2
2/14/1988	144	45.2	54.8
2/19/1988	148.5	28.7	71.3
12/17/03	74.6	25.3	74.7
01/19/05	85.1	33.7	66.3
01/22/05	77.8	32.5	67.5
01/01/08	85.9	39.5	60.5
02/09/08	96.7	24.3	75.7
02/27/08	95.5	39.4	60.6
02/21/09	117.6	32.0	68.0
01/02/10	101.3	27.7	72.3
01/29/10	104.0	36.0	64.0
02/13/10	92.0	33.8	66.2
1987-88 study average		25.0	75.0
This study average		32.4	67.6

A simple alternative method for estimating source attributions

Another estimate of crustal type sources (road dust and cinders) as percent of total PM was made by computing reconstructed fine soil (IMPROVE equation) and adding estimated associated carbonaceous aerosol mass ($EC + 1.8*OC$). Cinders have essentially no carbon, but road dust is about 10% OC and about 1% EC. As an upper limit it was assumed that OC and EC are proportional to the reconstructed soil at the same ratio as in road dust and this is added to the reconstructed soil calculation. The results (Table 10) show a higher average contribution from road dust and cinders (about 49%) than the CMB analysis (about 35%). Note: this method assumes organic and elemental carbon at the same abundance as in road dust and may be expected be an overestimate of carbon in road dust + cinders as cinders have essentially no carbon. It may thus represent an upper limit to the road dust and cinder contribution to PM10.

Table 10. Comparison of contribution of crustal elements from mass reconstruction calculations (soil+rd C) and CMB analysis (rd+cind).

date	soil+rd C	rd+cind%
12/17/03	35.9	27.1
01/19/05	48.2	36.3
01/22/05	44.7	35.0
01/01/08	62.0	42.5
02/09/08	31.3	26.1
02/27/08	56.2	42.5
02/21/09	52.8	34.2
01/02/10	52.9	29.6
01/29/10	54.7	38.6
02/13/10	53.9	36.6
average	49.3	34.8

Analysis of days with both PM10 and PM2.5 speciated data

Four days have both PM10 and PM2.5 speciated data. For these days, by subtraction of the PM2.5 attribution from the PM10 attribution we can get a value for PM coarse (PM10-PM2.5) attribution. These results are shown in Table 11.

Table 11. CMB attribution ($\mu\text{g}/\text{m}^3$) to wood smoke (FP) and road dust (rd+cind) by size fraction for four days with chemically speciated PM2.5 and PM10.

DATE	SIZE	stp conc	FP	rd+cind
12/17/03	10	74.6	53.0	19.7
12/17/03	2.5	40.6	28.6	1.5
12/17/03	coarse	34.0	24.5	18.3
1/13/05	10	39.3	31.1	5.4
1/13/05	2.5	32.7	27.5	0.6
1/13/05	coarse	6.6	3.6	4.7
1/19/05	10	85.1	51.8	29.5
1/19/05	2.5	30.5	28.1	2.2
1/19/05	coarse	54.6	23.7	27.3
1/22/05	10	77.8	47.5	25.5
1/22/05	2.5	33.2	27.6	1.9
1/22/05	coarse	44.6	19.8	23.6
average	10	69.2	45.8	20.0
	2.5	34.3	27.9	1.6
	coarse	34.9	17.9	18.5

Table 11 shows that over 90% of the PM10 mass attributed to road dust and cinders is in the coarse mode, as would be expected. However, on average for these days only a little more than 60% of the wood smoke (FP) is attributed to the fine mode, with about 40% in the coarse mode. This is somewhat unexpected, as a large majority of wood smoke is expected to be in the fine mode. The result is also contrary to the 1987-88 study which found the vast majority of the wood smoke impact in the fine mode. The attribution in the current study was largely a result of having a large residual in the reconstructed mass due to the inability to measure carbon on Teflon filters. The main question regards the “missing mass” which was assumed to be organic mass. If it is organic mass then what is it from if not wood smoke? Some would be associated with road dust, but not the levels needed. Some also would be from motor vehicles, but again this would almost all be expected to be in the fine mode. It is possible the reconstructed soil equation used is not appropriate for the local conditions and this would affect the residual used to estimate organic carbon. There could also be some water accounting for the missing mass, but concentration of hygroscopic compounds such as sulfate and nitrate are low and mainly in the fine mode, so this would not be expected to account for much. Because the wood smoke is present along with road dust, the road dust may become coated with wood smoke, thus causing a significant fraction of the wood smoke associated organic mass to be in the coarse mode.

Summary and conclusions

The question of the relative contributions of wood smoke and road dust to PM10 in Mammoth Lakes was considered. Teflon filters from high PM10 days between 2003 and 2011 were subjected to chemical analysis with XRF, ion chromatography, and filter light absorption. Four days analyzed also had PM2.5 filters that underwent chemical analysis. A major limitation was the inability to measure carbon on the filters. The filter light absorption provided a reasonable estimate of elemental carbon and the unexplained mass (on average 58% of PM10 and 68% of PM2.5) was assumed to be organic mass (OC*1.8). Estimates of contribution of wood smoke and road dust were made using Chemical mass balance (CMB) and a simple method based on abundance of crustal elements in the samples. For PM10 CMB showed an average of 32% due to road dust and 68% due to residential wood combustion. This compares to 25% from road dust and 75% from residential wood combustion for the winter 1987-88 study. The CMB maximum contributions (average of 3 highest days) to PM10 from road dust and residential wood combustion dropped by about 1/3 for each source category between 1987-1988 and 2008-2010.

A simpler method using reconstructed soil from the IMPROVE equation and adding organic mass and elemental carbon in the ratio found in Mammoth Lakes road dust gave an average of 49% contribution of road dust to PM10. There is likely some impact from other sources such as vehicle exhaust to the PM10 although these impacts are expected to be small in comparison to wood smoke and road dust. The relative contribution of wood smoke may have decreased since the 1990 AQMP was produced. The extent of any such decrease cannot be reliably determined with the data available. Additional measurements, chemical analysis and data analysis would be necessary to gain more confidence in the results.

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