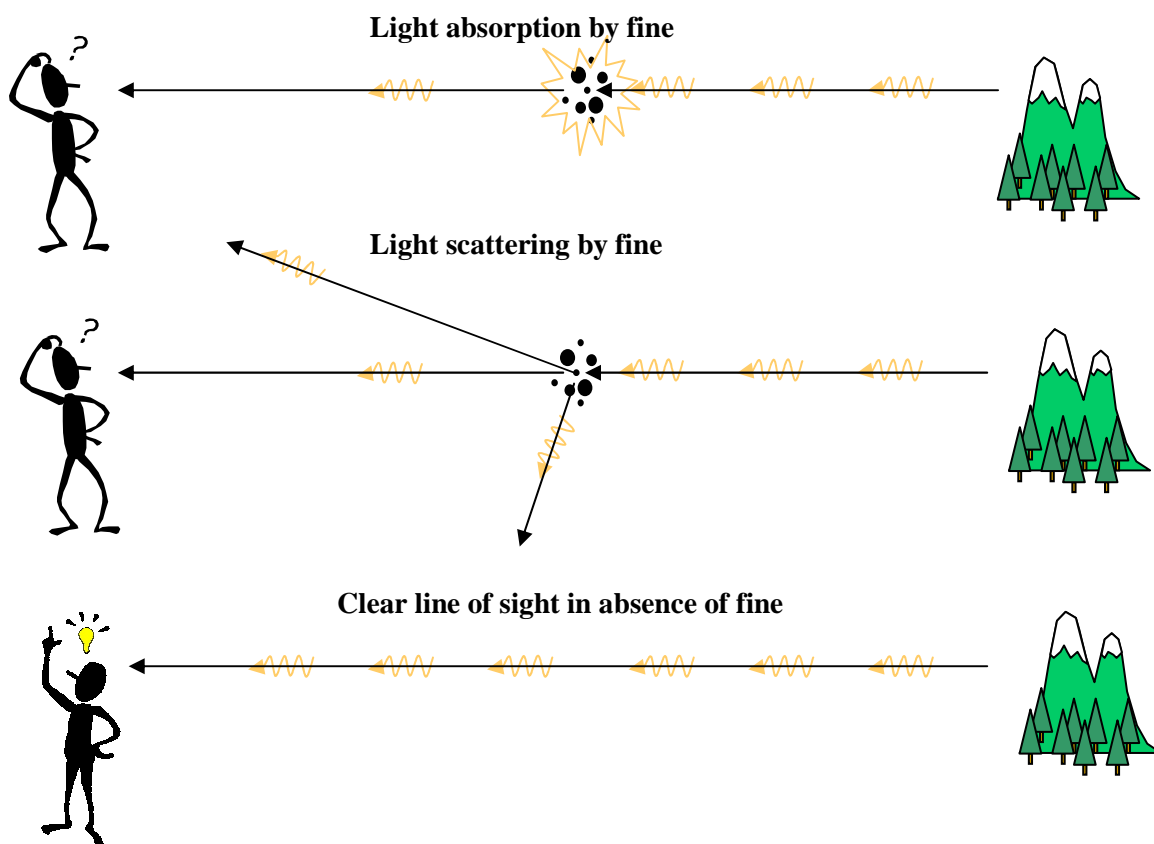


Haze Communication using the CAMNET and IMPROVE Archives: Case Study at Acadia National Park



Light extinction is a function of the individual light absorption and light scattering properties of particles and gases present in the atmosphere. The figure above illustrates absorption and scattering of light by particles. With a clear line of sight, all of the incoming photons, as represented by the squiggly yellow lines, arrive unimpeded to the eye of the observer. Absorbing particles take up some of the incident photons, whereas scattering particles divert the path of photons.

Total light extinction is frequently expressed as a light extinction coefficient (b_{ext}) in units of inverse length (such as Mm^{-1}). In simple terms, the light extinction coefficient is a measure of the proportion of light lost per unit of distance traveled through the atmosphere. The light extinction coefficient, b_{ext} , can be measured directly with a

transmissometer or determined empirically by “reconstructing” extinction as the sum of the scattering and absorption coefficients of the relevant particle constituents. The National Park Service’s IMPROVE program developed the following representation for reconstructed extinction:

$$b_{\text{ext}} = b_{\text{SO}_4} + b_{\text{NO}_3} + b_{\text{OrgC}} + b_{\text{Soil}} + b_{\text{Coarse}} + b_{\text{ElemC}} + b_{\text{Ray}}$$

where the subscripts refer the aerosol component. (SO₄ is sulfate, NO₃ is nitrate, OrgC is organic carbon, soil is fine soil, coarse is coarse mass, elemC is elemental carbon and Ray is Rayleigh scattering. Note Rayleigh scattering, b_{Ray} , is a measure of scattering due to air molecules. The Federal Land Managers’ Air Quality Related Values Workgroup (FLAG) uses a Rayleigh scattering value of 10 Mm⁻¹ for the entire U.S. (FLAG, 2000). This value corresponds to Rayleigh conditions at about 1800 meters above sea level (Sisler and Malm, 2000). However, Rayleigh scattering varies with altitude and at sea level is estimated to be about 12 Mm⁻¹ (Trijonis et al., 1990). It is understood that this reconstructed light extinction formula is not accurate for every sample, as it was designed to provide a consistent and replicable process for approximating light extinction based on observed relationships.

The calculation of extinction coefficients for each individual chemical species can be described by the following equations (FLAG, 2000):

$$b_{\text{SO}_4} = 3[(\text{NH}_4)_2\text{SO}_4]f(\text{RH})$$

$$b_{\text{NO}_3} = 3[\text{NH}_4\text{NO}_3]f(\text{RH})$$

$$b_{\text{OrgC}} = 4[\text{OrgC}]$$

$$b_{\text{Soil}} = 1[\text{Soil}]$$

$$b_{\text{Coarse}} = 0.6[\text{Coarse}]$$

$$b_{\text{ElemC}} = 10[\text{ElemC}].$$

IMPROVE assumes that all sulfate is in the form ammonium sulfate ((NH₄)₂SO₄) and that all nitrate is in the form ammonium nitrate (NH₄NO₃).

The bracketed quantities represent ambient air concentrations expressed in micrograms per cubic meter (µg/m³). The numeric coefficients represent “dry” scattering efficiencies (m²/g), while the relative humidity adjustment factor $f(\text{RH})$ accounts for the hygroscopic properties of sulfate and nitrate (i.e. their tendency to absorb water in the atmosphere). As relative humidity increases this factor becomes larger, which in turn produces a higher coefficient of light extinction for the hygroscopic particles. Provided concentrations and humidity levels are known, the light extinction coefficients for individual particle constituents can be calculated and summed to estimate the overall light extinction coefficient, b_{ext} .

Keep in mind the above equations reflect simplified assumptions about the role of relative humidity and may not adequately account for the non-linear relationship between

humidity and particle growth rate. Under the Regional Haze regulations, monthly average values based on climatology are used for the relative humidity factor. To better represent visibility extinction based on daily PM averages, corresponding daily relative humidity values should be used. However, some uncertainty is introduced due to diurnal variations in both RH and PM constituent masses. Further uncertainty is introduced by assuming all sulfate particles are fully ammoniated, since this is not always the case; more acidic particles have a higher affinity for water uptake and would require a corresponding larger $f(RH)$. Additionally, the above equations assume that organic carbon particles are non-hygroscopic and do not require a relative humidity adjustment. Whether or not a relative humidity adjustment factor should be applied to the organic fraction is an issue of current debate (Saxena et al., 1995) and is dependent upon the molecular makeup of the particles. Finally, the IMPROVE formulation was derived under the an assumption that the particles are externally mixed as opposed to the more likely case that each particle is a homogenous (internal) mixture of the individual components. This difference will affect the physical properties of the particle and in turn how they impact visibility. The sensitivity of reconstructed light extinction to each of these assumptions is an area that warrants further investigation.

The CAMNET photo-archive has been developed to demonstrate the variations in visibility conditions that exist at Acadia. Using 24-hour average measured values of speciated particulate mass and relative humidity, the archive attempts to explain how variations in these measured quantities affect local visibility conditions. The photographs display current/representative visibility conditions at Acadia NP for worst, average and best days (High, Middle, Low days). The discussion assists the reader in investigating the interactions of relative humidity and fine mass on visibility and provides a means to compare the visibility impact of individual species.

The first figure summarizes a range of visibility conditions for each of the four seasons. The next four pages of figures present the associated mass and visibility conditions. Discussions of these seasonal portraits are provided.

Text Descriptions for the Seasonal Figures:

Spring: Three days were chosen based on the total aerosol extinction to represent the three regimes (Best, Middle, Worst) of visibility at Acadia during the spring. Analysis of the mass composition shows that the distribution among the various aerosol constituents are similar across visibility category, with the hygroscopic sulfate and nitrate components accounting for 62-70% of the fine mass. Organic and elemental carbon accounted for 18-26% of the mass. For these days, both mass and relative humidity differences contribute significantly to the predicted visibility differences. Mass differences between the worst and middle day account for the visibility differences, whereas differences in humidity account for the differences between the middle and best days.

Based on the reconstructed extinction, clear differences should be discernible between all three cases. The worst day clearly shows the greatest impairment, while differences between the middle and best day are less obvious from the snapshots. Visibility degradation on the worst day is clearly dominated by higher mass values, with its humidity factor also playing a role. Differences between the middle and best summer day are predicted to be driven by differences in relative humidity. However, the 24-hour average $f(RH)$ factor calculated for the middle day is substantially greater than the actual factor associated with the humidity levels depicted in the photograph since the early hours of the day experienced very high relative humidity. The comparison of these two pictures illustrates the difficulty in combining 24-hour integrated or average measurements with instantaneous photographic results.

Summer: Three days were chosen based on the total aerosol extinction to represent the three regimes (Best, Middle, Worst) of visibility at Acadia during the summer. Analysis of the mass composition shows that the distribution among the various aerosol constituents are somewhat varied across visibility category, with the hygroscopic sulfate and nitrate components accounting for 44-66% of the fine mass. Organic and elemental carbon accounted for 19-39% of the mass. For these days, both mass and relative humidity differences contribute significantly to the predicted visibility differences, with the relative humidity effects dominating. In fact, the mass loading of the middle day is less than that of the best, but the relative humidity factor is almost 3 times as large.

Based on the reconstructed extinction, clear differences should be discernible between all three cases, and indeed, the worst day clearly shows the greatest impairment, followed by the middle day and then the best. The differences between the middle and best day is dominated by differences in $f(RH)$, although the difference in mass also plays a role. Visibility degradation on the worst day is clearly dominated by high relative humidity, in addition to the relatively greater percentage of mass attributable to hygroscopic species ammonium sulfate and nitrate. Humidity levels on this day are sufficiently high that visibility could be diminished due to fog. Differences between the middle and best summer day are driven again primarily by differences in relative humidity and mass composition, with both higher humidity and hygroscopic mass on the middle day. These

effects outweigh the fact that 15% greater mass was measured on the best day, relative to the middle day.

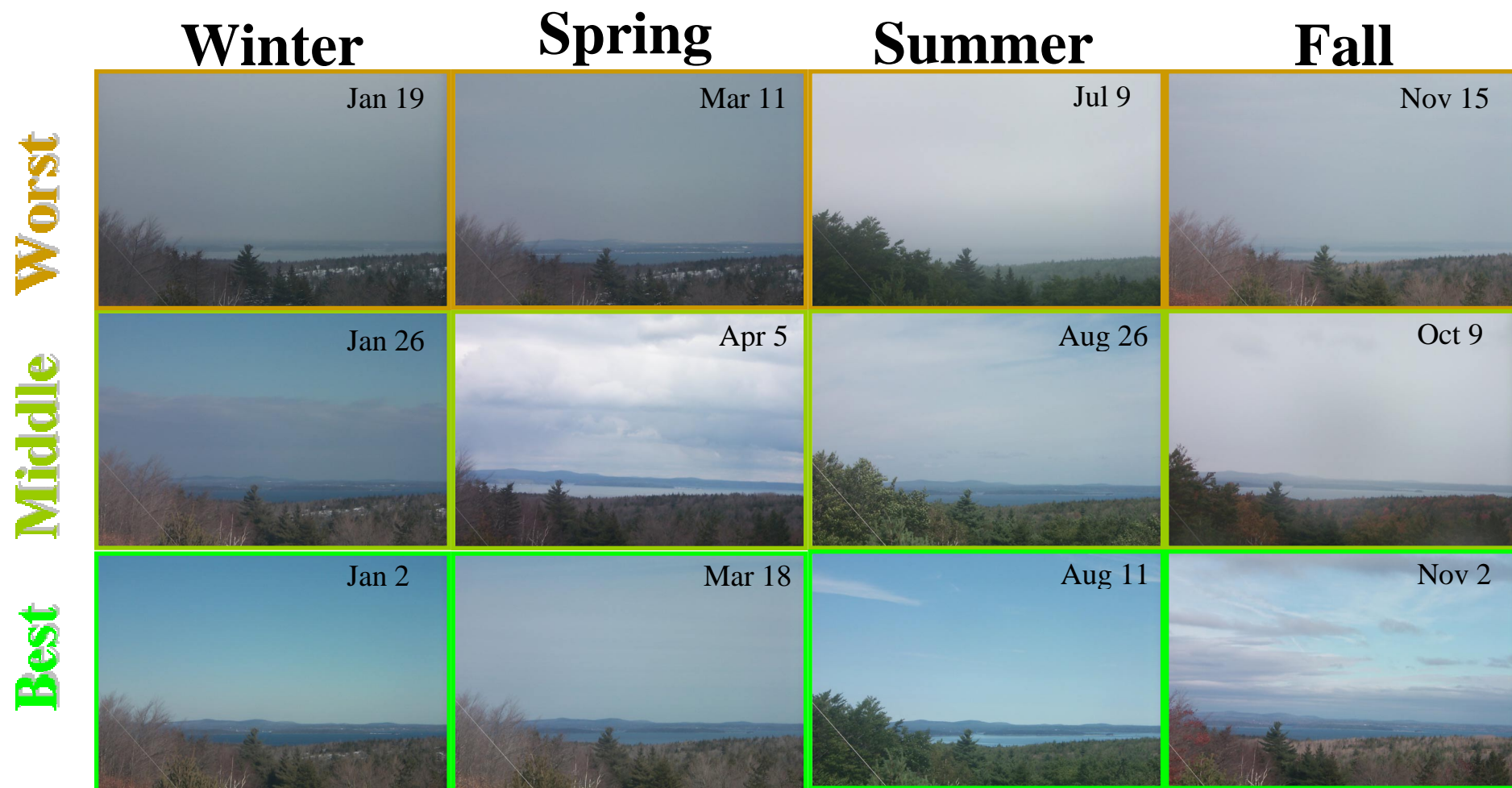
Fall: Three days were chosen based on the total aerosol extinction to represent the three regimes (Best, Middle, Worst) of visibility at Acadia during the fall. Analysis of the mass composition shows that the distribution among the various aerosol constituents are fairly similar across visibility category, with the hygroscopic sulfate and nitrate components accounting for 56-60% of the fine mass. Organic and elemental carbon accounted for 37-42% of the mass. For these days, both mass and relative humidity differences contribute significantly to the predicted visibility differences, with the higher mass values associated with greater visibility extinction. The relative humidity effect contributes over twice the punch to the middle day than it does to the best or worst day.

Based on the reconstructed extinction, clear differences should be discernible between all three cases, and indeed, the worst day clearly shows the greatest impairment, followed by the middle day and then the best. The differences between the middle and best day is dominated by differences in $f(RH)$, although the difference in mass also plays a role. Visibility degradation on the worst day is clearly dominated by the large observed particulate mass, more than 7 times the mass of the other days. This excessive particle loading more than outweighs the lower relative humidity effect on the worst day relative to the middle day.

Winter: Three days were chosen based on the total aerosol extinction to represent the three regimes (Best, Middle, Worst) of visibility at Acadia during the winter. Analysis of the mass composition shows that the distribution among the various aerosol constituents are fairly similar across visibility category, with the hygroscopic sulfate and nitrate components accounting for 54-63% of the fine mass. Organic and elemental carbon accounted for 30-40% of the mass. For these days, both mass and relative humidity differences contribute significantly to the predicted visibility differences, with the higher mass and $f(RH)$ values associated with greater visibility extinction.

Based on the reconstructed extinction, clear differences should be discernible between all three cases. The worst day clearly shows greater impairment than do the other two days. The differences between the middle and best photos are subtle. The pictures capture one moment of the day, whereas the filter measurements and calculations of extinction are integrated over an entire 24-hour period. There is a factor of 2 difference in mass and 50% difference in associated $f(RH)$. However, at the instance of the photographs, only a 10% difference in $f(RH)$ was observed. Actual instantaneous mass differences are not available, but may be considerably less than the 24-hour averaged difference suggests. These observations highlight the difficulties in combining real-time photographs with time-integrated measurements.

20% **Worst**/**Middle**/**Best** Visibility at Acadia (April 2000 to September 2002)

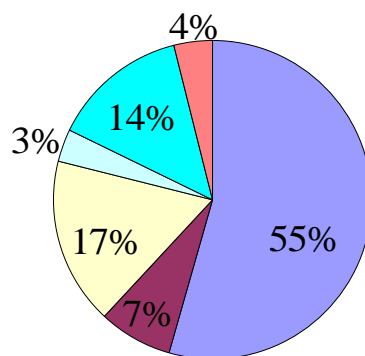
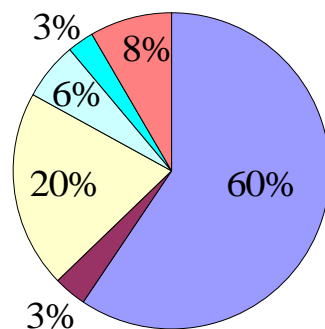
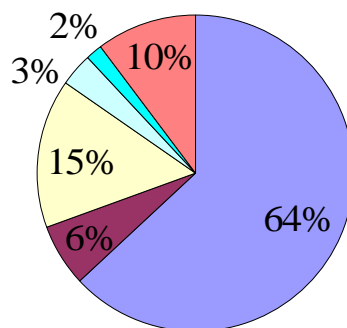


Worst
Middle
Best

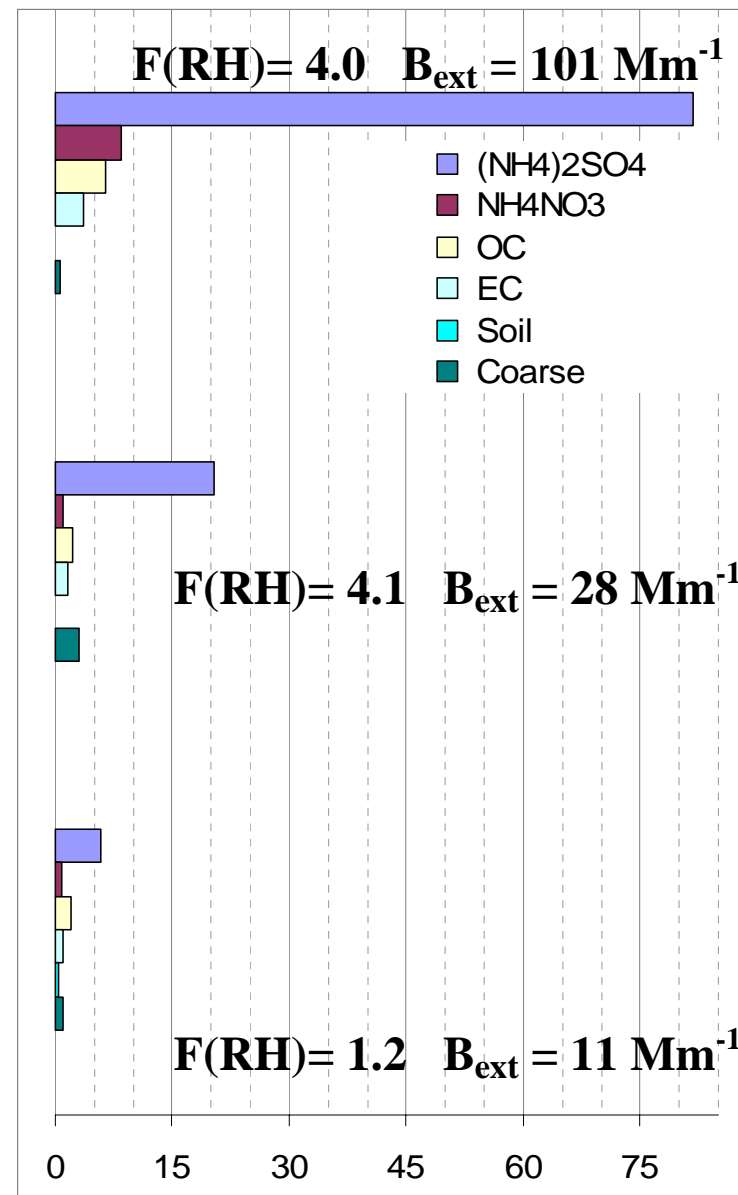
Spring



PM_{2.5} Mass Composition



Visibility Extinction (Mm⁻¹)

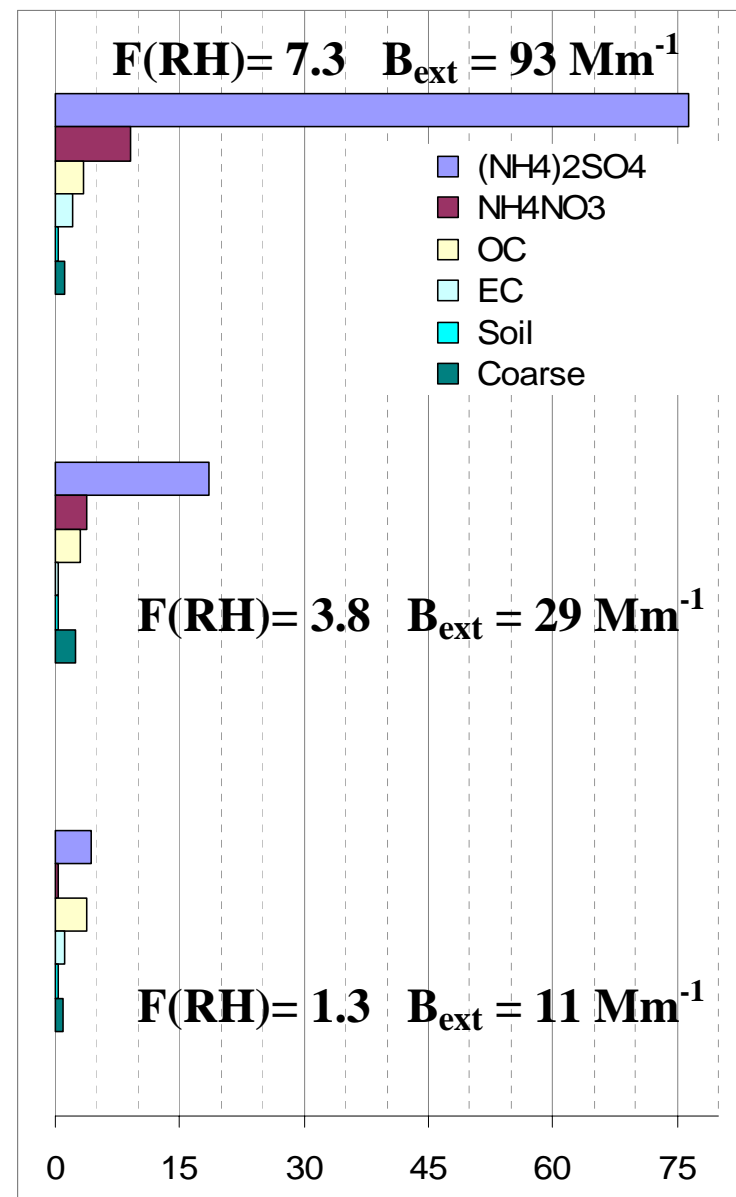
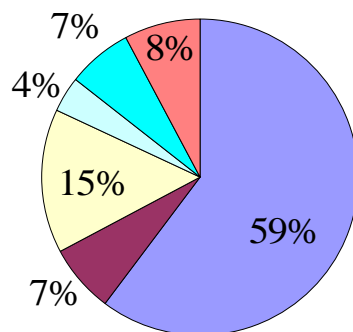


Summer

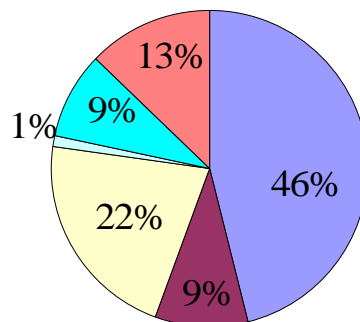
Mass Composition

Visibility Extinction (Mm^{-1})

Worst

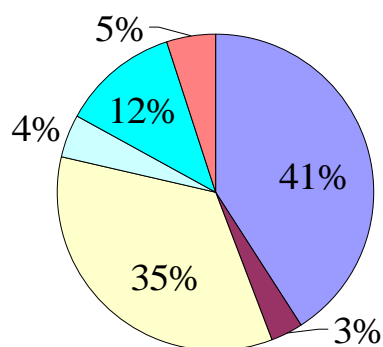
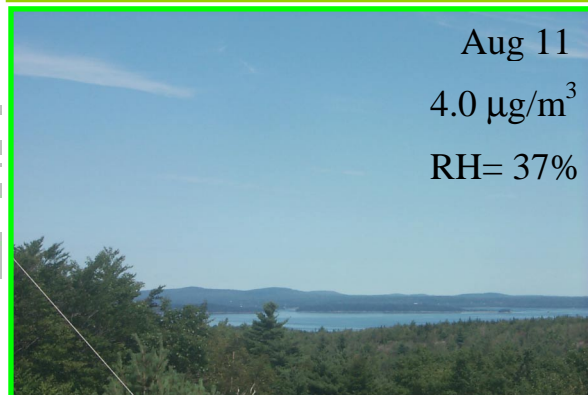


Middle



F(RH)= 3.8 B_{ext} = 29 Mm⁻¹

Best



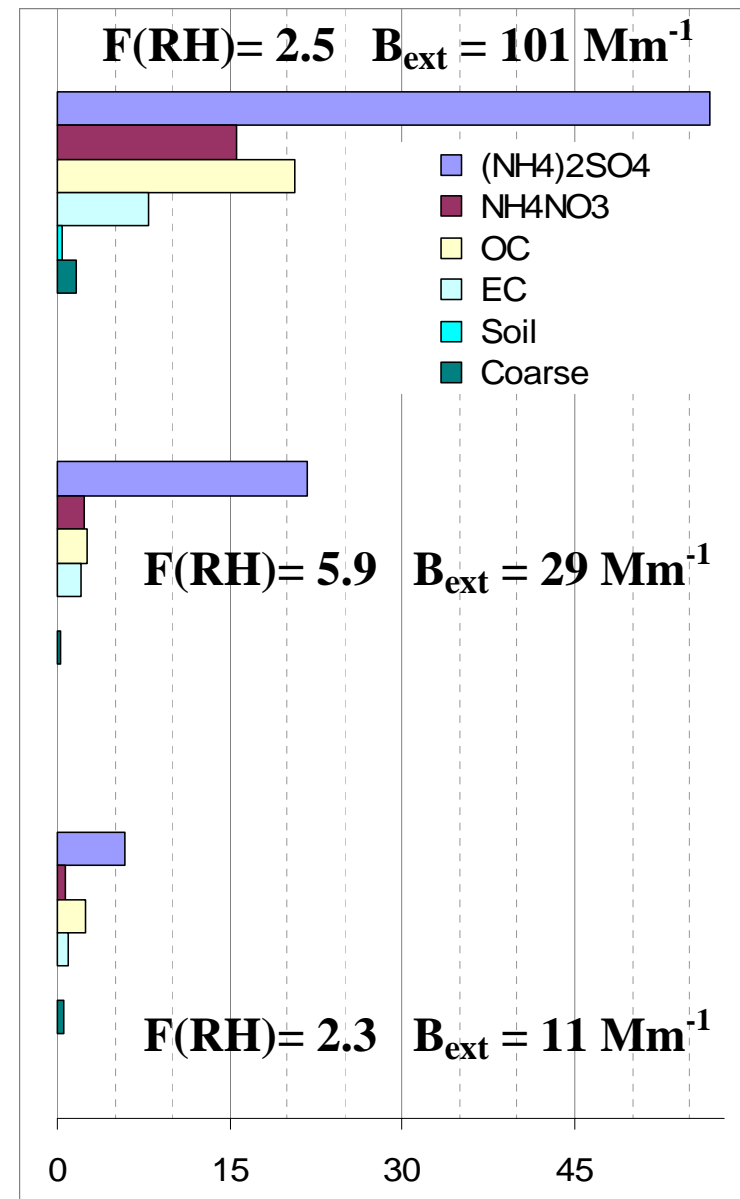
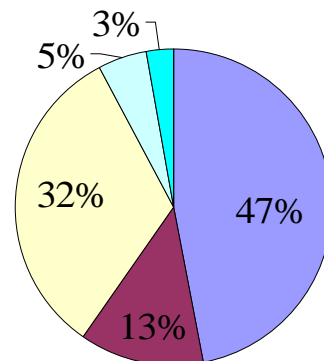
F(RH)= 1.3 B_{ext} = 11 Mm⁻¹

Fall

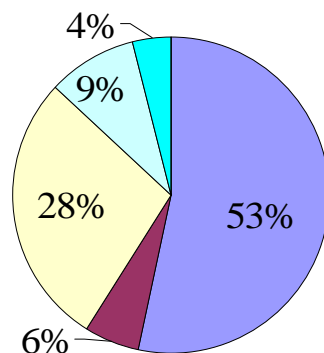
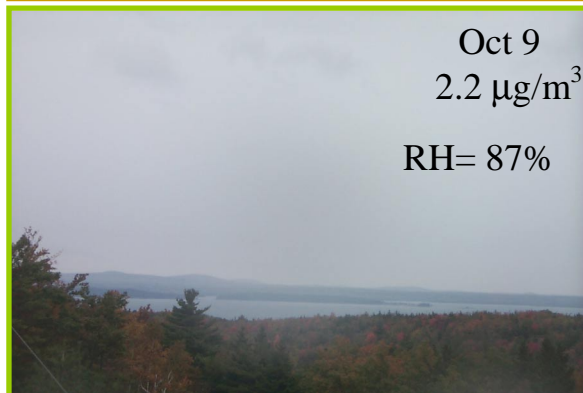
Mass Composition

Visibility Extinction (Mm^{-1})

Worst

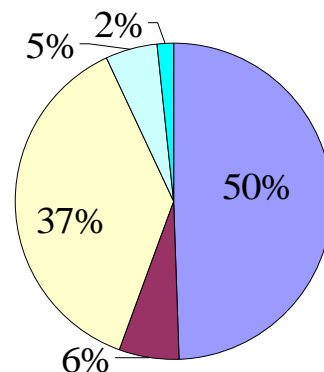


Middle



F(RH)= 5.9 B_{ext} = 29 Mm⁻¹

Best



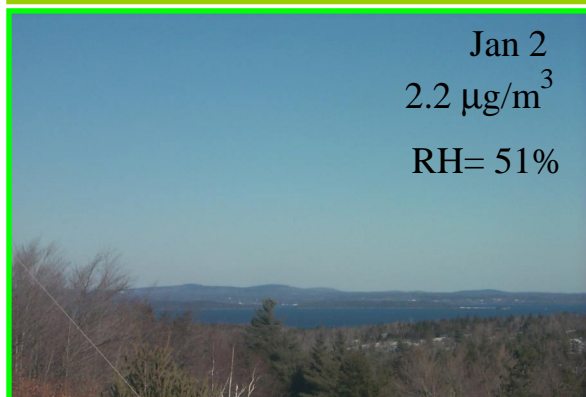
F(RH)= 2.3 B_{ext} = 11 Mm⁻¹

Worst

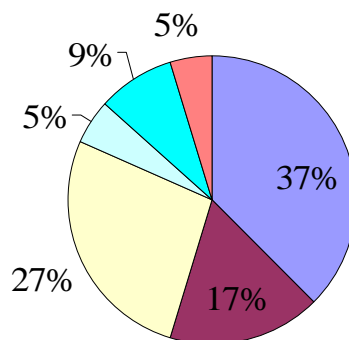
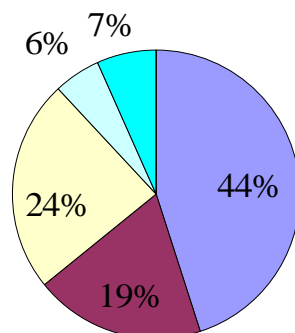
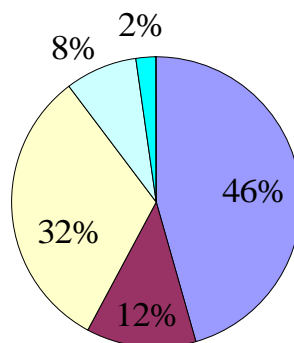
Middle

Best

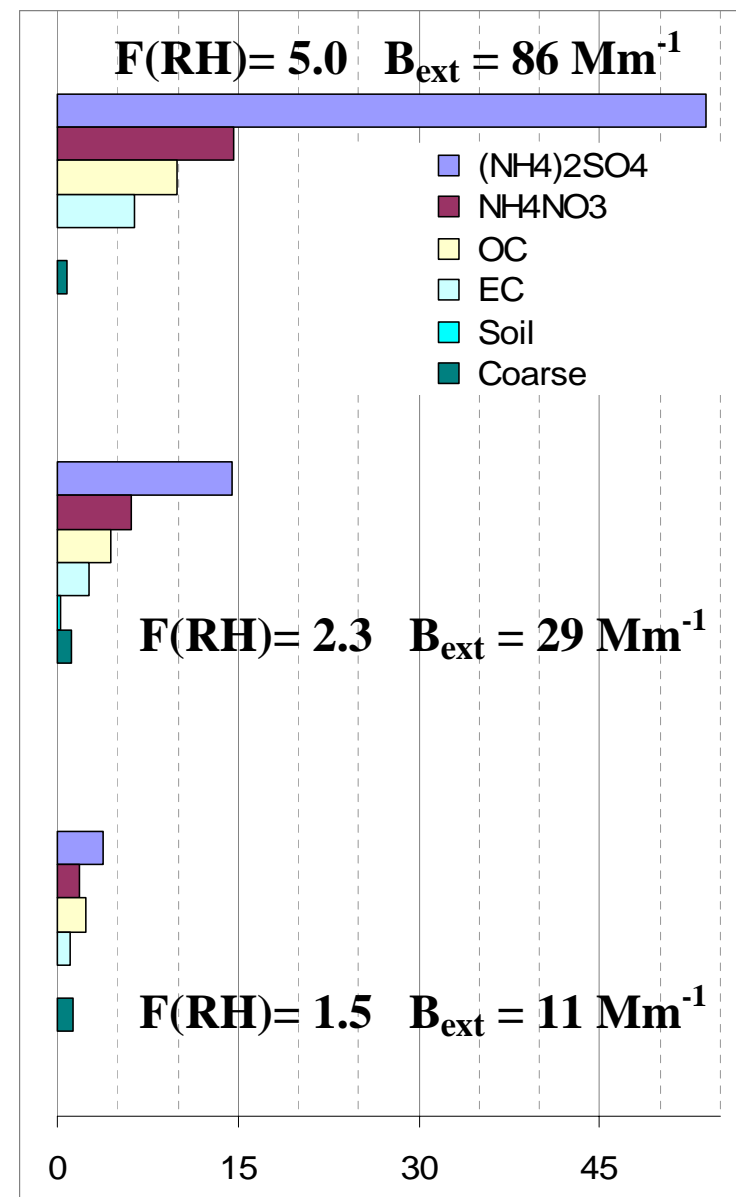
Winter



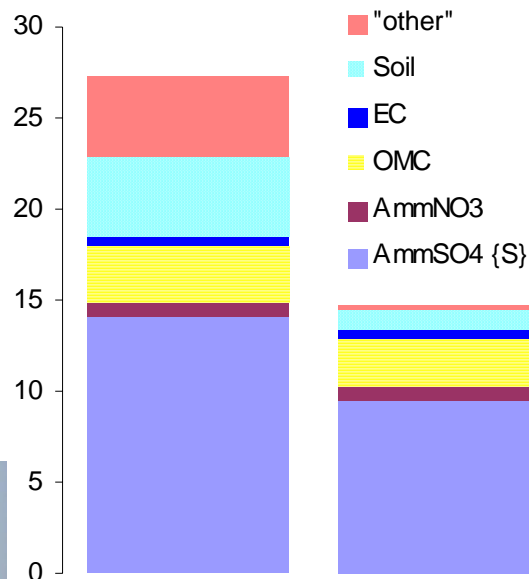
Mass Composition



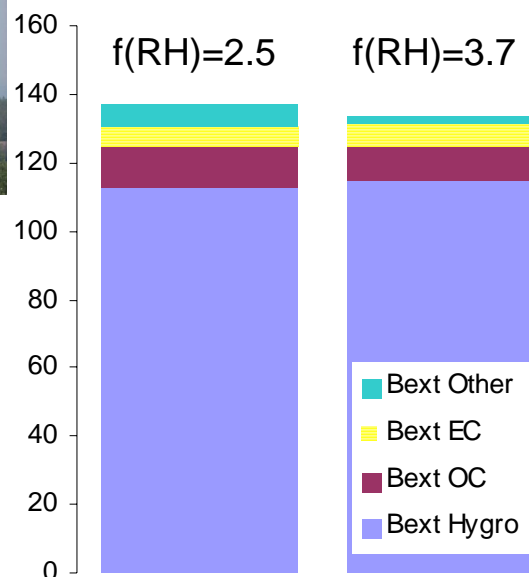
Visibility Extinction (Mm^{-1})



August 15, 2002

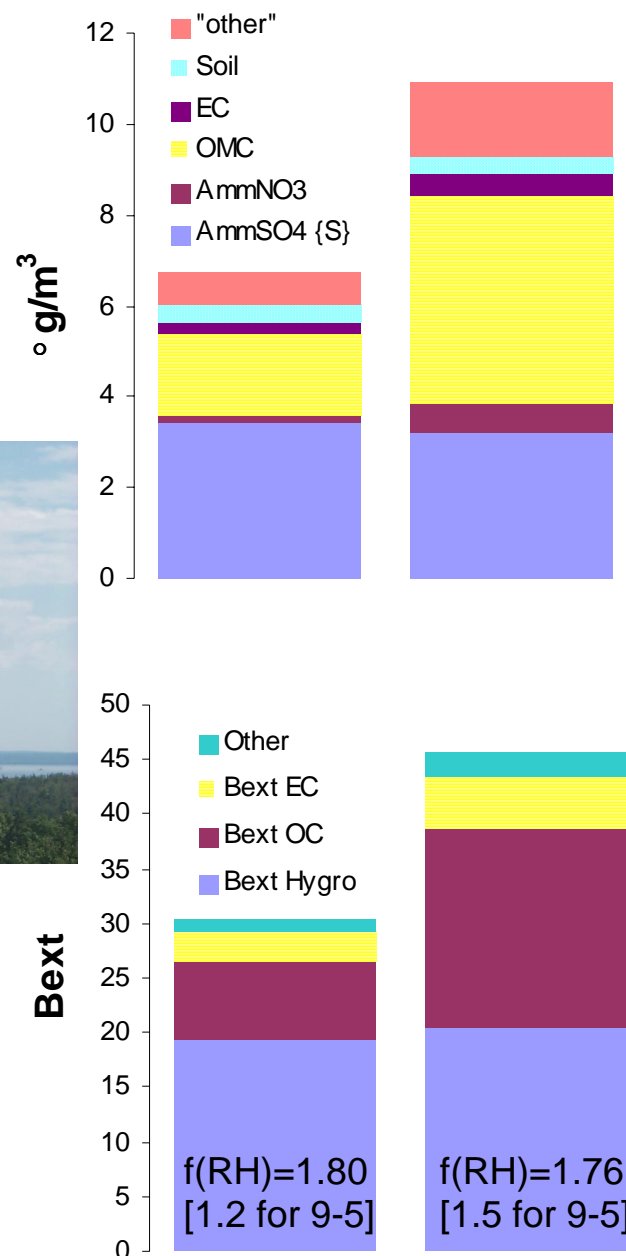


September 11, 2002



Two summertime days are compared. The visibility is quite poor for both days, based on the IMPROVE formula. Given the instantaneous photo versus the 24-hour average visibility calculation, this may not be the reality of the two pictures. Regardless, the main point to note here is that the particulate levels in the left figure are almost twice those in the right. However, due to higher humidity experienced on the day in the right side photo, the visibility is equally poor. This shows that even moderate pollution levels can lead to extremely poor visibility if the humidity is high and the level of hygroscopic particle constituents is appreciable.

July 30, 2001



July 13, 2002



Two summertime days are compared. The visibility is moderate for both days, based on the IMPROVE formula. Given the instantaneous photo versus the 24-hour average visibility calculation, the true extinction may be different. Here, visibility is slightly worse in the right-hand photograph. Based on the measured values, this is likely due to higher organic carbon levels measured on that day. The contribution due to hygroscopics is similar on both days. Note that the f(RH) values are similar when averaged over the 24-hour period, but in reality are slightly higher for the right-hand picture at the time of the photos. Since the hourly f(RH) is lower than the daily average (which is used in the bar charts), the bar chart may overestimate the relative contribution of the hygroscopics. Since we do not know how the actual levels of OC and sulfate at the time of the photo, the true visibility contributions cannot be determined.

The main idea in the final two pictorial presentations is to show that humidity levels, particle levels, and particle constituents all play a role in determining the visibility conditions. This effort also demonstrated the drawback of comparing 24-hour particle and humidity measurements with instantaneous photographs. With the new RAIN network collection hourly measurements of sulfate and bi-hourly levels of carbon, a more fruitful comparison with the CAMNET photos can be conducted. This may prove highly valuable in clarifying the roles of humidity and fine particle make-up on visibility conditions.