Regional Haze Assessments with CALPUFF: Application of Refined Procedures

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ABSTRACT

In 2001, the Federal Land Managers initiated a set of new procedures to assess the impact of proposed new sources on PSD Class I areas. These procedures, referred to as FLAG (Federal Land Managers' Air Quality Related Values Workgroup), often cause the assessment of regional haze impacts to be the most constraining issue with the new FLAG guidance. The guidance appears to be very restrictive in the following areas:

- The natural background extinction levels omit certain components, such as naturally occurring sea salt.
- The worst-case visibility impacts often occur during cloudy nighttime hours when there is no visibility AQRV.
- The high relative humidity (RH) periods are the most constraining, with FLAG requiring the applicant to consider RH up to 98%. However, such periods are often associated with precipitation events (which should be disqualified from consideration because of natural obscuration to visibility), but the FLAG guidance as implemented in CALPUFF does not currently allow the disqualification or special handling of precipitation cases.
- The daily average change in visibility impact due to a proposed source can be different depending upon how one does the averaging. The FLAG procedure uses extinction for each hour, which tends to heavily weight the hours (often at night) with poor visibility. An alternative approach would convert the hourly extinctions to visual range, thus emphasizing in the average (and appropriately so) the high visibility hours.

The paper is an update from one presented in 2002 by some of the authors, and it discusses these issues and how refinements in the modeling assessment approach, can be incorporated into the analysis. The authors also introduce newly available sources of meteorological data that have the capability of improving the accuracy of plume trajectories in CALPUFF modeling.

INTRODUCTION

In December 2000, the Federal Land Managers' Air Quality Related Values (AQRV) Workgroup (FLAG) issued a final Phase I Report[1]. FLAG consists primarily of representatives from the three Federal Land Managers (FLMs) that administer Federal Class I areas (U.S. Forest Service, National Park Service, Fish and Wildlife Service) supplemented with representatives from other vested groups, such as the Bureau of Land Management and the Environmental Protection Agency. The goal of FLAG is to provide consistent policies and processes in identifying and evaluating AQRVs for the review of new sources of air pollution. The FLAG Phase I Report consists of recommended procedures for FLMs to follow in the permit application process and specific guidance for the identification of AQRVs related to visibility, ozone and deposition.

The finalization of the FLAG Phase I guidelines was announced in the *Federal Register* on January 3, 2001. These guidelines have a significant effect upon one particular Air Quality Related Value, regional haze, and have significantly increased the challenge of permitting new, low-emission facilities, as reported by Paine et al.[2].

This paper is lengthy, so the authors present here a roadmap for the areas covered in the paper in the order that they appear.

1) FLAG regional haze assessment procedures are described, with particular attention to a) the use of 24-hr average concentrations, the role of relative humidity in the assessment, and natural background conditions.

2) The use of the CALPUFF model in long-range transport modeling is briefly discussed.

3) Technical issues involving the regional haze analysis procedures and their resolution are then discussed at length. There are several sub-sections: a) inclusion of sea salt in the determination of natural conditions, b) characterization of background visual range during precipitation events, and c) how daily averages of the source/background extinction ratio are determined in CALPUFF.

4) New sources of meteorological data input to CALPUFF are described.

FLAG REGIONAL HAZE ASSESSMENT PROCEDURES

Procedures are established in the final Phase I FLAG guidance report by which the FLM determines whether a proposed facility causes visibility impairment or contributes to a condition of pre-existing visibility impairment. The first step is to determine whether a source is to be evaluated in terms of the potential existence of a visible plume or whether it should be evaluated in terms of general haze. Plume visibility is a condition where a plume (or layered pollution) is discernable when viewed against a background sky or terrain on the background horizon. Haze is a condition where the plume becomes sufficiently well-mixed that the chief contribution is a reduction in visual range.

FLAG provides a rule of thumb that facilities within 50 km of a protected visibility area should be evaluated according to visible plume impact and that facilities beyond 50 km should be evaluated in terms of the contribution to haze. This paper addresses the more common case in which the proposed facility is more than 50 kilometers from the nearest PSD Class I area.

FLAG adopts the Interagency Workgroup on Air Quality Models (IWAQM) Phase II recommendations[3] on how to evaluate the contribution of a facility to general haze. This involves the application of the CALPUFF[4] model to estimate maximum 24-hour average concentrations of primary and secondary particulate. The modeled concentrations are then multiplied by an extinction efficiency that estimates the effect on absorption and scattering of visible light and then a relative humidity factor that simulates enlargement due to droplet formation on hygroscopic particles. The total modeled light extinction is then compared to a background extinction value to determine if the impact is significant. In making this comparison, FLAG inherently assumes that the modeled concentration at a single location is representative of a wide area surrounding the observer in the Class I area.

Use of 24-hr Average Concentrations at Peak Point Locations

Haze refers to a condition where the visual range is reduced in all directions over a wide region, while dispersion model predictions are available for individual receptor points and generally not for regions. Large changes in visual range commonly take place over a short time period (much less than a day), for example, with the passage of a front and the associated change in air mass. However, the monitoring data for particulate constituents that are used to reconstitute the background visibility are available only for daily averages. Because of the limitations of the available modeling and measurement tools, the IWAQM procedures have established that regional haze modeling assessments will be conducted *on a daily average basis at a single worst-case receptor location* so that the model predictions can be matched with the time period of the observations. The apparent rationale for the simplified IWAQM approach is that the use of a longer 24-hour averaging time instead of tracking hourly changes in visual range compensates, in part, for the use of a maximum point concentration to represent a lower spatial average concentration.

It is also well known[5,6,7] that air quality models cannot predict accurately at a specific time and location for short-range models, much less long-range models. The failure to achieve this goal is due in part to turbulent eddies or very small wind circulations that cannot be measured or even modeled because the observational and data input requirements are too great. These turbulent eddies cause slight plume trajectory deviations close to the source that forever after alter the trajectory of a plume, and make good model performance paired in time and space an virtually unattainable goal. Therefore, any attempt to try to specify 1-hour visibility impacts at a particular location from a specific source is problematic due to this model limitation. IWAQM's use of a 24-hour average concentration as an accurate estimate of the average plume-related concentration within an observer's line-of-sight (i.e., from a point in the Class I area out to a distance equal to the background visual range) tends to avoid a dependence on very short-term model estimates, which are uncertain for a specific time and location. Essentially, the IWAQM procedures use temporal averaging as a surrogate for spatial averaging.

One important aspect of the use of a 24-hour average is that it includes the period between dusk and dawn, a period when visibility is not typically an important AQRV. This is especially true for cloudy nights when no celestial objects can be viewed. Furthermore, the nighttime hours are most often associated with the highest relative humidity near the ground due to nocturnal cooling. The high nighttime relative humidity causes the growth of hygroscopic particles and therefore results in the highest regional haze impact when visibility is naturally restricted because of darkness. This issue is discussed in more depth below.

Relative Humidity Extinction Adjustment

The relative humidity adjustment that is used to compute plume-related extinction is a major contributor to the worst-case predictions of regional haze impacts. Moisture plays an important role because particles that are amenable to condensation nuclei sites for water vapor will form small droplets at relative humidity values less than 100%. These enlarged "particles" are then much more efficient at scattering light than dry particles. For values of relative humidity (RH) approaching 100%, the scattering efficiency can increase by a factor as high as 18 (at 98% RH in CALPOST) over the dry case. Therefore, periods of high relative humidity will often lead to the worst-case regional haze impact predictions. IF EPA's guidance on tracking progress under the regional haze rule limits the f(RH) to 95% RH, then CALPOST applications should use the same RH cap.

The use of average relative humidity as measured at surface stations (as recommended by FLAG) can be biased for the purposes of visibility impairment because periods of very high relative humidity generally do not correspond to weather conditions during which visibility is an AQRV. For example, visibility is not an important AQRV during periods of rain, snow, or fog when visibility is naturally reduced. Such natural reductions in visibility are not currently considered under FLAG guidance in the characterization of "natural conditions" and they are not currently accounted for in the CALPOST post-processing procedures that compute visibility impacts of a proposed source. Also ignored by FLAG is the influence of naturally occurring sea salt particles on the reduction of visibility, especially in humid coastal areas. This omission, which is discussed in more detail below, can substantially increase the conservatism of regional haze impact analyses for coastal locations. In general, the FLAG and IWAQM procedures do not address reduced natural visibility during periods of increased humidity and often ascribe the largest modeled visibility impacts to periods of complete darkness or significantly reduced visibility due to fog or precipitation.

Natural Background Conditions

FLAG uses the maximum 24-hour modeled concentration of primary and secondary particulate, adjusted by mean relative humidity, to estimate the extinction associated with emission sources. This value is then compared to the natural background extinction for the Class I area that is listed in Appendix 2.B of the FLAG report. As noted previously, the natural background extinction is intended to represent the state of the atmosphere in the absence of human activity, based on the 1990 NAPAP report[8]. Table 2.B-2 of that Appendix lists the presumed constituents of the natural background. For the continental United States, it corresponds to an annual PM_{10} concentration of about 5 ug/m^3 in the Eastern United States and 4 ug/m^3 in the Western United States. These values are a small fraction of the levels that have been characteristic of many of the Eastern Class I areas since the system of National Parks and wilderness areas was established in the early 20th century.

FLAG suggests the following criteria by which the FLM will develop recommendations: if there is no pre-existing haze concern, a single PSD source must not have impacts that exceed 5% of the natural background. If the source impacts exceed 5%, a cumulative analysis must demonstrate that the impact of all PSD sources combined does not exceed 10% of the natural background. If the pre-existing haze cumulative analysis has already established that combined

PSD impacts exceed 10% of the natural background, a facility may contribute no more than 0.4% of the natural background extinction.   Although commenters on the FLAG guidance suggested that these thresholds are overly protective, FLAG rejected these comments.

The authors consider that it is inherently incorrect to consider the total absence of anthropogenic emissions as a "natural condition" because this is inconsistent with the presence of civilization. Air emissions that result from human activities date back to the Stone Age when fire was first used for cooking and space heating. Up until the beginning of the 20th century, Native Americans had used fire for centuries for various purposes[9], including

- Hunting
- Crop management
- Improvement of vegetation growth and yields
- Reduction of fire-prone “fuels” (prescribed burning)
- Insect collection
- Pest management
- Warfare & signaling
- Economic extortion
- Clearing areas for travel
- Felling trees
- Clearing riparian areas.

Since the beginning of the 20th century, fire suppression has resulted in a buildup of vegetative “fuels” and catastrophic wildfires. Recent estimates of background visual range, such as NAPAP[8] may have underestimated the role of managed fire on regional haze. Various government agencies are now planning to increase prescribed burning to reduce the threat of dangerous wildfires. The increased presence of the atmospheric loading of particulate due to burning needs to be included in background visual range estimates attributed to “natural conditions”. While this adjustment is not further discussed in this paper, it is yet another factor that makes the present estimates of natural background visual range excessively high. In addition, natural biogenic emissions of volatile organic compounds need to be included in the estimates of natural conditions.

USE OF CALPUFF FOR LONG-RANGE TRANSPORT MODELING

PSD permit applicants should use a steady-state plume model for distances up to 50 kilometers. For longer distances, CALPUFF is recommended. The FLMs require a PSD Class I assessment to be conducted for all proposed sources within 100 kilometers of a PSD Class I area. For distances between 100 and 200 kilometers, some sources with very low emissions may be exempted from PSD Class I considerations. Major sources with emissions well in excess of 250 tons per year of SO_2, NO_2, and/or PM_{10} will likely be required to conduct a modeling assessment if the source is within 200 or even 300 kilometers of a PSD Class I area. As noted by Paine et al.[2], and Walcek[10], and Moran and Pielke[11], the inability for CALPUFF to account for wind shear effects on additional plume dispersion produces a plume that is too compact, and limits the ability of CALPUFF to provide unbiased predictions beyond 200 kilometers or 12 hours of transport time.

Because the conservative screening CALPUFF procedures may show significant impacts from even low-emission proposed projects, most applicants will likely need to conduct a refined modeling analysis with full CALMET processing, as noted in the Wygen 2 project in Wyoming[12]. This occurs because the significant impact thresholds are only 4% of the PSD Class I increments for SO, NO_2, and PM_{10}, making it potentially difficult for a project to show insignificant impacts. The effort required to conduct a refined analysis is substantial.

In practice, one of the most daunting aspects of a refined PSD Class I analysis is obtaining a valid and complete background source inventory. Many state inventories are in poor condition, if they exist at all, and some states (such as New York) require the applicant to obtain verification in writing from each background source facility for every exhaust parameter input value being modeled. The effort to acquire a background emission inventory can take up to several months and significant cost to complete.

A number of CALPUFF runs conducted were conducted by Paine et al.[2] to determine the air quality impact of a hypothetical well-controlled coal-fired source. Their analysis indicated that the most restrictive aspect involves the regional haze analysis. Although the hypothetical project also showed significant impacts for SO_2 increment consumption, a cumulative analysis may resolve the increment consumption issue because the increment significance level is only 4% of the allowable total. However, with the regional haze cumulative impact threshold set to only 10% for all sources combined (just twice the significance level for only the proposed source), it is clear that this element of the analysis is often the controlling one. Therefore, much of the focus of this paper is on CALPUFF regional haze modeling.

TECHNICAL ISSUES INVOLVING THE REGIONAL HAZE ANALYSIS PROCEDURES AND THEIR RESOLUTION

The tendency for CALPUFF modeling results of regional haze impacts to dominate the permitting process for new sources with the implementation of FLAG guidance has resulted in increased scrutiny of the shortcomings of the FLAG guidance and the modeling procedures involved. Paine et al.[2] described several technical problems with the way the prescribed system is set up to analyze regional haze impacts. This paper updates this discussion and offers specific enhancements to CALPUFF to help eliminate the shortcomings.

1) Inclusion of Sea Salt in "Natural Conditions" Depiction of Background Visual Range

In the Draft Guidance for Estimating Natural Visibility Conditions Under the Regional Haze Program[13], there is a discussion in Section 1.11 regarding the preliminary estimates of natural conditions. This discussion notes that the NAPAP report[8] from which the estimates are derived "provides annual average estimates of natural concentrations of these six main components of PM for eastern and western regions of the country." These estimates were used to estimate natural background under the FLAG guidance. The six components referred to in the quotation are sulfate, nitrate, organic carbon, elemental carbon, and crustal materials. Sea salt is not included in the list. Furthermore, since the estimates in the NAPAP report are averages over the entire eastern and western parts of the country, they do not include the influence of sea salt at coastal and near-coastal locations.

Because Class I areas, especially those on or near ocean coastlines, might have significant contributions from naturally occurring sea salt aerosols, the lack of their inclusion may significantly underestimate the natural background light extinction. This discussion presents and documents example estimates of the average contributions of sea salt aerosols to light extinction in coastal and near-coastal Class I areas in the Southeast United States. The same procedure can be used for any PSD Class I area, but the largest effect will be realized for PSD Class I areas near the ocean.

The contribution to light extinction by a specific aerosol component is typically expressed as:

$$E = k\, f(RH)\, [\text{component}] \qquad (1)$$

where:

E = contribution to light extinction by the specific component (Mm^{-1})

k = light extinction efficiency of the component at low relative humidity (also called the "dry" light extinction efficiency) (m^2/g)

f(RH) = an empirical function describing the increase in light extinction due to the growth of particles of a hygroscopic component as the relative humidity (RH) increases

[component] = atmospheric concentration of component ($\mu g/m^3$)

The following steps were used to estimate the sea salt contributions using Equation 1:

1. Annual and seasonal average sea salt aerosol concentrations ([Sea Salt]) at one coastal and one near-coastal Class I area in the Southeast were estimated using data collected by the Interagency Monitoring for the Protection of Visual Environments (IMPROVE) program.

2. The technical literature was reviewed to estimate the dry light extinction efficiency (k) and the variation of light extinction by sea salt aerosols [f(RH)] with relative humidity.

3. Equation 1 was applied to the annual and seasonal average sea salt aerosol concentrations to estimate annual average contributions to estimate contributions to light extinction.

Sea Salt Aerosol Concentrations

Sea salt aerosol concentrations were estimated from sodium and chloride concentrations measured at IMPROVE network monitoring sites, based on the assumption that all of the sodium and chloride are present in naturally occurring sea salt. Data for Cape Romain National Wildlife Refuge (NWR), on the coast of South Carolina, and for Okefenokee NWR, near the coast of Georgia, were used. Measurements at Cape Romain NWR began in early-September 1994, and measurements at Okefenokee NWR began in late-September 1991. Data are available from the IMPROVE Web site[14].

The IMPROVE database includes reported $PM_{2.5}$ concentrations of elemental sodium, ionic chloride and elemental chlorine. Seasonal averages of the reported values of sodium and chloride were calculated. The chlorine data were not used, because chlorine is volatilized from the filter during sampling[15]. The definitions of the seasons followed the definitions used by IMPROVE: winter is December, January and February; Spring is March, April and May; summer is June, July and August; and fall is September, October and November. Concentrations below the reported method detection limit (MDL) were set to one-half the MDL prior to calculating the average values. As shown below, a substantial number of values were available for each season, so no substitutions for missing data were made. The annual average concentrations were calculated as the averages of the four seasonal average concentrations. This averaging of the seasonal averages avoided biases introduced by uneven distributions of available data among seasons.

The average concentrations are presented in Table 1, along with the ratio of average sodium to average chloride. The ratio of sodium to chloride in seawater is about 0.56 (Gartrell et al.[16]), while the ratios in the table all exceed 2.0. As described by Tang et al.[17], this chloride deficiency can be caused by reactions with sulfuric or nitric acid that liberate gaseous hydrogen chloride and increase concentrations of sulfate or nitrate in the sea salt particles. Chloride deficits in sea salt particles may also be caused by reactions with gaseous nitrogen dioxide or by oxidation of dissolved sulfur dioxide by ozone.

Gartrell et al.[16] used the percentage of sodium in sea salt to estimate the atmospheric concentration of sea salt prior to chloride loss. They then assumed that the lost chloride was replaced by sulfate (one sulfate ion for two chloride ions) to estimate the "aged" sea salt concentration after chloride loss. This approach leads to a higher mass concentration of sea salt aerosol than would be present if the chloride were not displaced, because the formula weight of sulfate is larger than the atomic weight of chlorine. To allow for the possibility that this process occurs under natural conditions, this same approach was used to estimate aged sea salt concentrations from the average sodium concentrations in Table 1. The following equation was used for the calculation:

$$[\text{Aged Sea Salt}] = [\text{Na}] / 0.306 + 1.35\,(1.79\,[\text{Na}] - [\text{Cl}^-]) \quad (2)$$

where:

[Aged Sea Salt] = Sea salt concentration ($\mu g/m^3$)

[Na] = Sodium concentration ($\mu g/m^3$)

0.306 = Mass fraction of sodium in sea salt (Gartrell et al.[16])

1.35 = Formula weight of sulfate (96) divided by two times the formula weight of chloride (35.5)

1.79 = Mass ratio of chloride to sodium in sea salt

[Cl^-] = Chloride concentration ($\mu g/m^3$)

The last term in Equation 2 accounts for the displacement of chloride by sulfate.

Because chloride displacement by other substances may not occur under natural conditions, when concentrations of acidic gases and particulate constituents would be lower, a lower-limit estimate for the sea salt concentration was calculated by using only the first term in Equation 2.

Table 1. Seasonal and annual average sodium, chloride and chlorine concentrations.

		Sodium		**Chloride**		
Site	**Season**	**Concentration ($\mu g/m^3$)**	**Number**	**Concentration ($\mu g/m^3$)**	**Number**	**Sodium/ Chloride**
Cape Romain NWR	Winter	0.380	140	0.173	128	2.20
	Spring	0.518	118	0.142	106	3.65
	Summer	0.388	120	0.134	107	2.90
	Fall	0.308	140	0.104	130	2.96
	Annual	0.398		0.138		2.88
Okefenokee NWR	Winter	0.215	220	0.084	187	2.56
	Spring	0.316	207	0.111	170	2.85
	Summer	0.271	189	0.091	158	2.98
	Fall	0.243	195	0.117	165	2.08
	Annual	0.261		0.101		2.58

Values are based on IMPROVE monitoring data.

The resulting estimates of the seasonal and annual average aged sea salt aerosol concentrations are listed in Table 2. The lower and upper limits of the estimated annual average concentrations are about 1.3 $\mu g/m^3$ and 2.1 $\mu g/m^3$, respectively, at Cape Romain NWR, and about 0.9 $\mu g/m^3$ and 1.4 $\mu g/m^3$, respectively, at Okefenokee NWR. For comparison, the estimated annual average natural concentration in the East of hygroscopic $PM_{2.5}$ constituents proposed in US EPA Draft Guidance for Estimating Natural Visibility Conditions Under the Regional Haze Rule[13] is 0.33 $\mu g/m^3$, which is about a third or less of the aged sea salt aerosol mass concentration estimates.

Table 2. Seasonal and annual average estimates of aged sea salt aerosol concentrations.

Site	Season	Lower Limit[a] (μg/m^3)	Upper Limit[b] (μg/m^3)
Cape Romain NWR	Winter	1.242	1.926
	Spring	1.692	2.751
	Summer	1.269	2.026
	Fall	1.006	1.609
	Annual	1.302	2.078
Okefenokee NWR	Winter	0.703	1.110
	Spring	1.032	1.645
	Summer	0.887	1.419
	Fall	0.795	1.225
	Annual	0.854	1.350

[a] Lower limit assumes no replacement with chloride by other substances (i.e., "fresh" sea salt aerosol).
[b] Upper limit assumes chloride replacement by sulfate.

Sea Salt Aerosol Light Extinction Efficiency

Information regarding the dry light extinction efficiency for sea salt particles was not found in the technical literature. However, the dry light extinction efficiency is generally related to the size distribution of the particles at low relative humidity, although other factors such as refractive index also play a part. Gartrell, et al.[16] have shown that the typical particle size distributions for soil and for sea salt are very similar. The dry light extinction efficiency, for fine soil is commonly accepted to be around 1 m^2/g (Malm, et al.[18]). Therefore, 1 m^2/g was used as the dry particle light extinction efficiency for sea salt in these analyses.

The hygroscopic nature of salt particles is well established (Tang[19], Tang and Munkelwitz[20], Tang, et al.[17], Ansari and Pandis[21]). Both pure salts (e.g., NaCl) and mixed salts (e.g., KCl-NaCl) have been shown to exhibit substantial particle growth as a function of relative humidity. Sea salt particles often contain organic materials in internal mixtures, and these mixed salt-organic particles have been shown to be hygroscopic, as well (Ming and Russell[22]). Furthermore, the hygroscopic properties of salt particles are generally similar to those of ammonium sulfate and ammonium nitrate (the hygroscopic species represented by f(RH) values in Table 2.A-1 of the FLAG Phase I Report[1]). For example, the deliquescence humidity (at 25° C) is 75.7% for NaCl, compared to 79.5% for ammonium sulfate (Tang[16]).

Specific values of f(RH) for sea salt have been determined through field measurements. Gasso, et al.[23] conducted aircraft-based measurements of the aerosol over the east subtropical Atlantic Ocean, near the Canary Islands. Their measurements were conducted in June and July of 1997 as part of the Aerosol Characterization Experiment 2 (ACE2). They used a humidograph, consisting of two nephelometers attached to the same inlet probe with a 2.5 μm cutpoint. One nephelometer measures ambient light scattering, and the inlet to the other nephelometer is heated to provide a measure of scattering by dry particles. This dual sampling approach measures two points on the scattering versus RH curve, in order to obtain an estimate of the dependence of aerosol light scattering on RH.

The ACE2 measurements obtained data in three classes of ambient conditions: polluted, dust, and marine. The marine days (no pollution or dust as determined by back trajectory modeling) represented light scattering by sea salt particles.

The marine days data yielded the following f(RH) function:

$$f(RH) = (1 - RH/100)^{-\gamma} \quad (3)$$

where:

$\gamma \quad = \quad 0.6276 \pm 0.1159$

When this equation is applied to RH, it yields numerical values of f(RH) as shown in Table 3. Also shown, for comparison, are the CALPOST f(RH) values from FLAG Table 2.A-1.

It is evident from Table 3 that the sea salt f(RH) values match those from CALPOST reasonably well, especially between 60% and 90% RH. Therefore, within reasonable uncertainty bounds, the CALPOST f(RH) values in FLAG Table 2.A-1 can be used to model the growth of sea salt particles.

Table 3. F(RH) for sea salt particles and for the CALPOST hygroscopic species (ammonium sulfate and ammonium nitrate).

RH (%)	f(RH) - Sea Salt	f(RH) - CALPOST
5	1.0	1.0
10	1.1	1.0
15	1.1	1.0
20	1.2	1.0
25	1.2	1.0
30	1.3	1.0
35	1.3	1.0
40	1.4	1.1
45	1.5	1.2
50	1.5	1.2
55	1.7	1.3
60	1.8	1.4
65	1.9	1.7
70	2.1	1.9
75	2.4	2.2
80	2.7	2.7
85	3.3	3.4
90	4.2	4.7
95	6.6	9.8

Contributions of Sea Salt Aerosols to Light Extinction

The FLAG guidance provides f(RH) values for use with seasonal and annual average concentrations of ammonium sulfate and ammonium nitrate (FLAG[1]). Table 2.B-1 of the guidance document lists these values for individual Class I areas along with estimates of the extinction coefficient for natural conditions. The values in Table 2.B-1, along with a dry light extinction efficiency of 1 m^2/g, were applied to the estimates of seasonal and annual average sea salt aerosol concentrations to estimate sea salt aerosol contributions to light extinction.

Estimated seasonal and annual average aged sea salt aerosol contributions to the light extinction coefficient are listed in Table 4, and the estimated total seasonal and annual average light extinction coefficients without and with the aged sea salt aerosol contributions are listed in Table 5. As seen in Table 5, including the aged sea salt aerosol contribution increases the estimated natural background light extinction coefficient significantly. The lower and upper bounds for the percentage increase in the annual average estimated light extinction coefficient are 20 and 32 percent, respectively, at Cape Romain NWR. The lower and upper bounds for the percentage increase in the annual average light extinction coefficient at Okefonekee NWR are 14 and 22 percent, respectively.

Table 4. Estimated seasonal and annual average aged sea salt aerosol contributions to light extinction.

Site	Season	f(RH)[a]	Concentration ($\mu g/m^3$)		Light Extinction Coefficient[b] (Mm^{-1})	
			Lower Limit	**Upper Limit**	**Lower Limit**	**Upper Limit**
Cape Romain NWR	Winter	2.9	1.24	1.93	3.6	5.6
	Spring	3.3	1.69	2.75	5.6	9.1
	Summer	3.9	1.27	2.03	4.9	7.9
	Fall	3.3	1.01	1.63	3.3	5.3
	Annual	3.3	1.30	2.08	4.3	6.9
Okefenokee NWR	Winter	3.2	0.70	1.11	2.3	3.6
	Spring	3.4	1.03	1.65	3.5	5.6
	Summer	3.9	0.89	1.42	3.5	5.5
	Fall	3.6	0.80	1.23	2.9	4.4
	Annual	3.5	0.85	1.35	3.0	4.7

[a] From FLAG [1], Table 2.B-1
[b] Based on 1 m^2/g dry light extinction efficiency

Table 5. Estimated seasonal and annual average light extinction under natural conditions with agaed lt aerosol contributions.

Site	Season	Light Extinction Coefficient Without Sea Salt[a] (Mm^{-1})	Aged Sea Salt Contribution to Light Extinction Coefficient (Mm^{-1})		Light Extinction Coefficient With Aged Sea Salt Contribution (Mm^{-1})	
			Lower Limit	Upper Limit	Lower Limit	Upper Limit
Cape Romain NWR	Winter	21.1	3.6	5.6	24.7	26.7
	Spring	21.4	5.6	9.1	27.0	30.5
	Summer	22.0	4.9	7.9	26.9	29.9
	Fall	21.5	3.3	5.3	24.8	26.8
	Annual	21.5	4.3	6.9	25.8	28.4
Okefenokee NWR	Winter	21.3	2.3	3.6	23.6	24.9
	Spring	21.5	3.5	5.6	25.0	27.1
	Summer	22.0	3.5	5.5	25.5	27.5
	Fall	21.7	2.9	4.4	24.6	26.1
	Annual	21.7	3.0	4.7	24.7	26.4

[a] From FLAG[1], Table 2.B-1

2) Characterization of Background Visual Range During Precipitation Events

The natural background assumed by FLAG ignores natural obscuration during fog and precipitation events. This is a major omission that has led to unnecessarily conservative estimates of proposed project impacts ever since FLAG was implemented in early 2001. Recently, the assistant secretary of the Department of the Interior, Mr. Craig Manson, in a letter dated January 10, 2003 regarding the Roundup Power Plant permit application in Wyoming, has carefully considered evidence that peak predicted impacts due to a proposed source occur during periods of natural obscuration. This concept should be made a permanent feature of the FLAG process. One way to do this is described below.

Currently, the relative humidity at the nearest surface station is used to adjust the natural background visual range (or extinction) due to the sensitivity of hygroscopic particulate to humidity. The same meteorological station (or site-specific measurements in the applicable PSD Class I area, as used by Pearson and Nall[24]) can be used to determine whether there is any fog or precipitation, and if so, what the visual range is. For hours with detected precipitation, the background visual range should be taken directly from measurements (airport visibility, or Class I areas transmissometer); this value would replace the natural background used during times of fog and precipitation. During periods at night when there is a cloud ceiling (coverage more than 50%), the natural background visual range would be set to zero.

These suggestions are generally in line with the points made by Dr. Warren White[25] in his comments on the Air & Waste Management Association's Critical Review of Visibility issues last year.

Dr. White explained in his review that the Regional Haze Rule overlooks other plausible ways to assess visibility degradation. For example as Dr. White notes, in California, the procedures for assessing visibility impacts have reasonable alternatives:

- Daytime visibility only is assessed (in this paper, we propose that nighttime visibility during periods of an observed cloud ceiling be assigned a background visual range of zero).
- Periods of elevated humidity are discarded from further review. White notes that IMPROVE optical measurements at relative humidities greater than 90% are withheld from summary calculations since they are deemed to be subject to "weather interferences". However, the FLAG guidance requires relative humidities as high as 98% to be included in regional haze calculations. That is why the authors propose a cap of 90% on relative humidity for CALPOST computations of visibility impairment.
- Visibility is characterized in terms of visual range, rather than particle extinction.

The authors agree with Dr. White and recommend the changes proposed in this paper as consistent with his recommendations.

3) Daily Averages of the Source/Background Extinction Ratio:

Paine et al.[2] discussed the effects of the FLAG guidance in permitting a well-controlled hypothetical emissions source in the Midwest. This discussion reviews part of that paper. In conjunction with Figure 1, we present in Table 6 the CALPUFF computed hourly light extinction from a source and the background, as well as hourly source/background ratios. FLAG and IWAQM require the computation of hourly light extinction because the source-related extinction is a function of the hourly particulate concentration, and both the source-related and background extinction are functions of the hourly relative humidity. Humidity affects only the fraction of particulate that is hygroscopic. Because source-related particulate (beyond 50 km) is primarily hygroscopic whereas "natural background" particulate is mostly non-hygroscopic, high humidity has a greater effect on source-related extinction than on natural background extinction. As noted previously, the use of the daily average extinction at a worst-case receptor rather than an hourly average extinction using a spatial average is tied to the availability of daily particulate measurements and the relative difficulty of computing spatial averages.

Figure 1. Example of Diurnal Trend of Relative Humidity and CALPUFF-Predicted Extinction Change

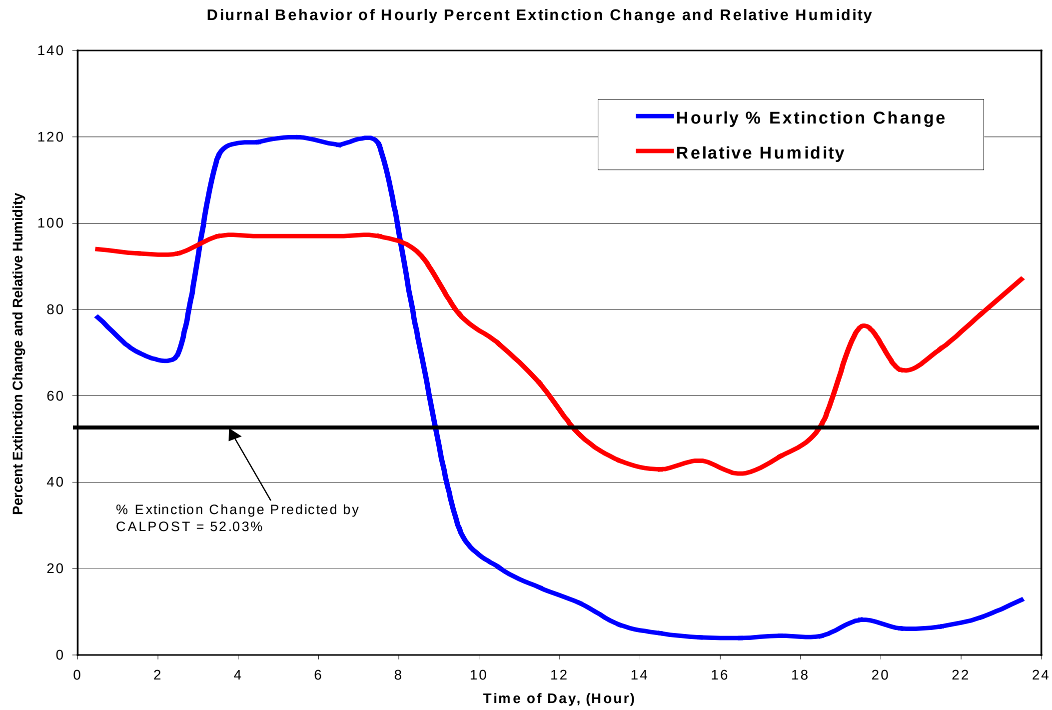


Table 6. Hourly light extinction ratio computations from enhanced CALPOST output.

HR	RH(FRAC)	F(RH)	BEXT(BKG)	BEXT (source)	Hourly Extinction Change	BKG Vis. Range (km)	Degraded Vis. Range (km)
0	0.94	8.371	26.034	20.354	0.7818	150.27	84.33
1	0.93	6.963	24.766	17.365	0.7012	157.96	92.85
2	0.93	6.963	24.766	17.292	0.6982	157.96	93.01
3	0.97	15.077	32.07	37.06	1.1556	121.98	56.59
4	0.97	15.077	32.07	38.106	1.1882	121.98	55.75
5	0.97	15.077	32.07	38.461	1.1993	121.98	55.46
6	0.97	15.077	32.07	37.888	1.1814	121.98	55.92
7	0.97	15.077	32.07	37.899	1.1818	121.98	55.91
8	0.93	6.963	24.766	17.886	0.7222	157.96	91.72
9	0.79	2.601	20.841	6.117	0.2935	187.71	145.11
10	0.72	2.008	20.307	4.109	0.2023	192.64	160.22
11	0.63	1.547	19.892	3.111	0.1564	196.66	170.06
12	0.51	1.237	19.613	2.358	0.1202	199.46	178.05
13	0.45	1.161	19.545	1.363	0.0697	200.15	187.11
14	0.43	1.144	19.529	0.983	0.0503	200.32	190.72
15	0.45	1.161	19.545	0.796	0.0407	200.15	192.32
16	0.42	1.134	19.521	0.77	0.0394	200.40	192.79
17	0.46	1.172	19.555	0.871	0.0445	200.05	191.52
18	0.53	1.267	19.64	0.859	0.0437	199.19	190.84
19	0.76	2.263	20.537	1.667	0.0812	190.49	176.18
20	0.66	1.699	20.029	1.235	0.0617	195.32	183.97
21	0.71	1.96	20.264	1.346	0.0664	193.05	181.03
22	0.79	2.601	20.841	1.833	0.0880	187.71	172.53
23	0.87	3.785	21.906	2.798	0.1277	178.58	158.35
			23.4269583	**12.188625**	**geometric mean:**	**173.16**	**138.02**
% Extinction Change that CALPOST predicted -				**52.03%**	**0.2046**	22.59	28.34
						AVG BEXT = ^	AVG BEXT = ^
					Ext. Change Using Degraded Vis. Range: 25.47%		

In the FLAG approach, the daily average source-related and background extinction values are computed separately as the arithmetic means of the computed hourly extinction values. The ratio of these mean values is computed daily and the largest of the daily ratios are used to evaluate the significance of a source's contribution to haze. The FLAG method is not a valid measure of the average visibility impairment for a number of reasons:

1) A few hours with very large high humidity tend to dominate the source-related average, thus biasing the daily ratio on the high side. This feature is evident in Figure 1, where the high humidity at night dominates the daily total and results in a reported average that visually appears to overestimate a representative average ratio for the day. In fact, the CALPUFF-reported average daily ratio of about 52% is higher than 15 of the 24 hours in the sample day, and is higher than most of the daylight hours.

2) The AQRV that has traditionally been associated with regional haze is the visual range (Malm[26]). For instance, a park visitor can readily notice whether distant mountains can be seen but cannot directly sense or relate to changes in extinction. Theoretically, visual range is defined as the distance that a dark object can be seen against the horizon sky. In a practical sense, the smaller the distance of a landscape feature to the visual range, the more clearly the feature can be seen. Thus, a visitor's enjoyment of a scenic vista and visual range are directly related. As noted, visual range varies from hour to hour, with changing air masses, humidity, weather conditions and pollution levels. In terms of protecting this AQRV, a pertinent question is a project's impact on the daily visual range. This can be determined by taking the average visual range for natural background conditions and then determining the change after the source's contribution is considered.

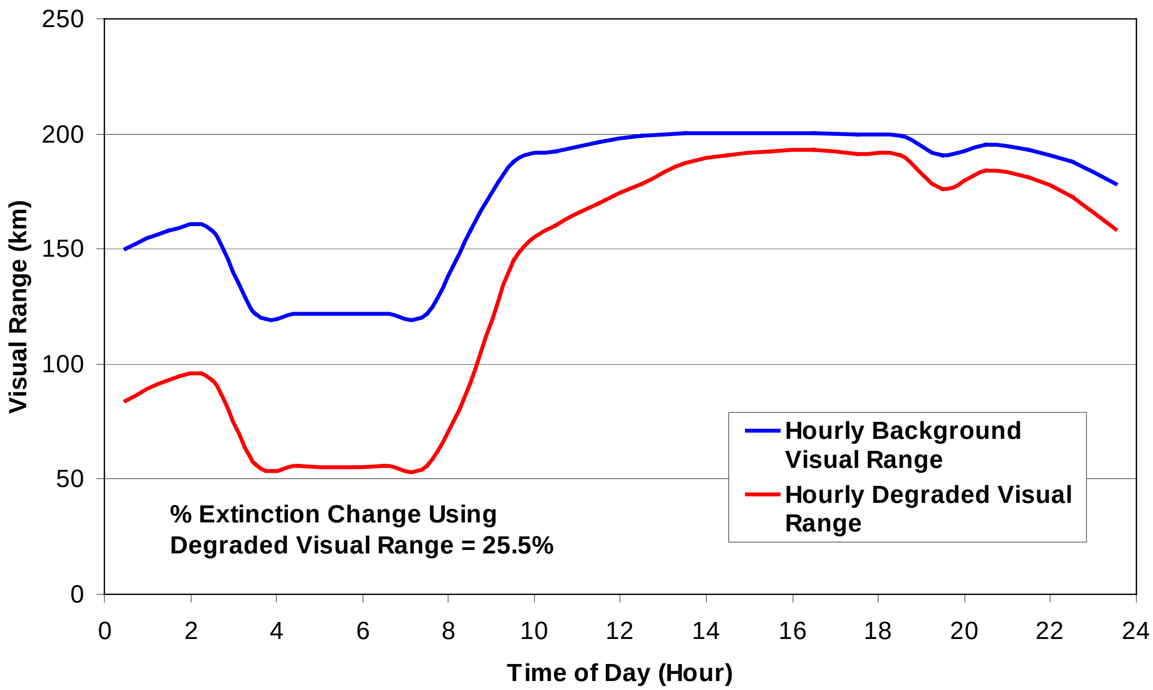
In the FLAG and IWAQM guidance, light extinction is used rather than visual range, in part, because it is easier to quantify light extinction (from particulate measurements) than visual range (Malm[26]). Assuming that the concentration is spatially uniform, at any moment in time the visual range is inversely proportional to the atmospheric extinction. However, because extinction is not constant during throughout the day but varies from hour to hour, the daily average visual range is not inversely proportional to the daily average extinction. The current CALPOST code computes hourly source and background extinction changes. It then separately computes the daily average source and background extinction and takes the ratio of these daily averages. However, this ratio does not relate to visual range during the important daylight hours, as shown in Figure 1, and therefore has little relevance in characterizing the park visitor's daily visual experience.

ENSR has enhanced the CALPOST code to report hourly extinction changes and visual ranges at the user's option. These hourly ratios are shown in the example in Table 2 that relates to Figure 1. As discussed above, a few nighttime hours dominate the daily average source-related extinction. The CALPOST averaging method allows an inappropriate dominance of this relative short nighttime period on the daily average extinction source/background ratio. To correct this problem, we recommend that CALPOST be modified to convert the light extinction for natural background conditions to visual range, the more direct representation of the visibility AQRV. This conversion is done using the Koschmieder relationship noted in the FLAG Phase I report[1] that the visual range in kilometers is related to the light extinction (b_{ext}, in inverse megameters) due to the light scattering by particles plus Rayleigh scattering:

Visual Range (km) = $3912/b_{ext}$.

This alternative method computes the change in the visual range before and after the addition of the source's contribution, which is a more relevant AQRV than changes in daily average extinction. The change in the visual range is illustrated in Figure 2.

Figure 2. Diurnal Trend of Visual Range With and Without Source Impact – Same Case as Shown in Figure 1



The proposed method to be used in computing the change in the light extinction due to the proposed source can be demonstrated in the spreadsheet shown in Table 2. In the table, the current CALPOST calculation of the extinction change is presented as the ratio of the daily averages of the source-related extinction to the natural conditions extinction, resulting in a 52% change. The use of an average of hourly ratios of the extinction change could be considered, with the use of a geometric mean to compute the daily average ratio as the appropriate mathematical averaging method (Giese[27]). However, zero CALPUFF predictions for some hours would complicate this method. What we consider to be a better method is shown in the columns in Table 2 labeled as background visual range and degraded background visual range. These last two columns show a computation of the hourly natural background visual range in kilometers (3912/bext) and also the "degraded" visual range (3912 divided by the sum of the natural background extinction plus the source-caused extinction). The table shows that we can then take a daily average visual range for both columns and then compute the corresponding *effective daily average light extinction* in both cases. The change in the extinction is then computed and compared to the 5% significance change threshold. In this case, the resulting change is about

25%, which is less than half of the CALPOST-reported change with the use of the same input data!

It is evident from Table 2 that different methods to average the results of the same CALPUFF model predictions can result in strikingly different answers. The use of the proposed "degraded visual range" method is, in our view, more compatible with the visual experience of a person in a Class I area who is cognizant of whether distant landscape features can be seen but does not directly experience extinction. We are, therefore, determining whether the change in the visual range is significant in terms of whether it can be detected. The CALPOST computation currently takes the average of the hourly light extinction due to the source emissions in the absence of any natural atmospheric components. This method, in our view, does not properly account for the incremental change in the visibility as well as the proposed degraded visual range technique.

Another benefit that results from the adoption of the degraded visual range technique is that individual hours with poor natural visibility (due to cloudy nighttime hours and/or periods of precipitation or fog) can be properly accounted for by the use of a zero visual range, consistent with the presumption that such conditions are excluded from natural conditions (Malm[26]). The visibility degradation due to the source would then be appropriately minimized, and those individual hours would not (and should not) substantially affect the daily average of the visual range and the light extinction change.

The proposed solution for this issue is to change CALPOST to use the degraded visual range method. For hours of cloudy conditions at night (more than 50% cloud cover) and/or periods of precipitation or fog, a modified natural background value for visual range would be used, as discussed earlier in this paper.

NEW SOURCES OF METEOROLOGICAL DATA INPUT TO CALPUFF

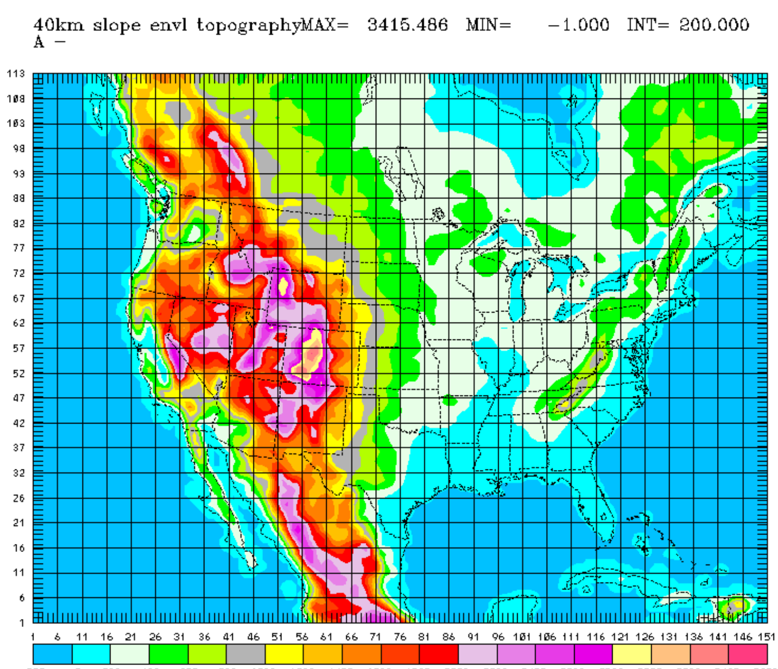
In CALMET, the use of MM5 prognostic wind field data is usually incorporated as a superior initial wind field estimate (in Step 1) prior to correction to actual observations (in Step 2). The Step 1 process also takes the initial wind field estimate and subjects it to refinements due to terrain effects and minimization of divergence (to preserve conservation of mass laws). The result of this Step 1 process is far superior to that using a crude initial wind field estimate, which then would require a substantial correction to observations in the Step 2 process. With the use of traditional observations (widely scattered airports and balloon sounding stations,), the CALMET Step 2 process needs to have a large radius of influence for the correction of a crude initial wind field estimate. This tends to smooth out the wind field relative to what is available as details in an MM5 data set. As an alternative, the use of the MM5 data for Step 1 is often associated with very local corrections in the Step 2 process. As noted above, IWAQM[3] has observed improved CALPUFF performance with the prognostic wind field model used as a Step 1 initial guess field.

Prognostic (predictive) models are well known to have significant advantages over diagnostic wind field models. Dynamic constraints are those resulting from the application of conservation laws involving time derivatives, such as conservation of momentum. The chief drawback of prognostic models is the computational expense of running them. Computational stability considerations require that the models be stepped forward with a time-step that is proportional to the grid cell size. Thus, high-resolution grids require an extremely large number of time-steps to be computed in order to cover the needs of a long-term air quality study. For this reason, high-resolution prognostic models are most often applied to episodic case studies.

While the application of customized prognostic meteorological models to long-term air quality studies can in some cases be prohibitively expensive, data from the National Oceanographic and Atmospheric Administration (NOAA) prognostic model outputs and analyses can be combined with mesoscale data assimilation systems to produce high-resolution data sets of long duration. NOAA runs a suite of models at varying initial times, resolutions, domains of coverage, and forecast duration. Each model run starts with results from a previous run, combined with all available observed data, including surface and upper air observations, satellite, and radar data. This process of combining the various data sources to yield a unified representation of the three-dimensional atmosphere is termed assimilation.

Assimilation has been an area of active research over the years. As increasingly accurate analyses become available, combining more data types is one of the principal means for improving forecast quality. A promising data archive for air quality applications is NOAA's RUC2 model data. RUC2, or Rapid Update Cycle Version 2, is a short-term forecast model that is re-initialized each hour based on previous model results and actual meteorological readings. The RUC2 model[28] grid contains 40 km cells, with over 40 layers of data in the vertical dimension (see Figure 3). This resolution is sufficient to easily represent the upper air features captured by the rawinsonde network. Interested parties, including private companies, can download the RUC2 model data from a NOAAPORT server.

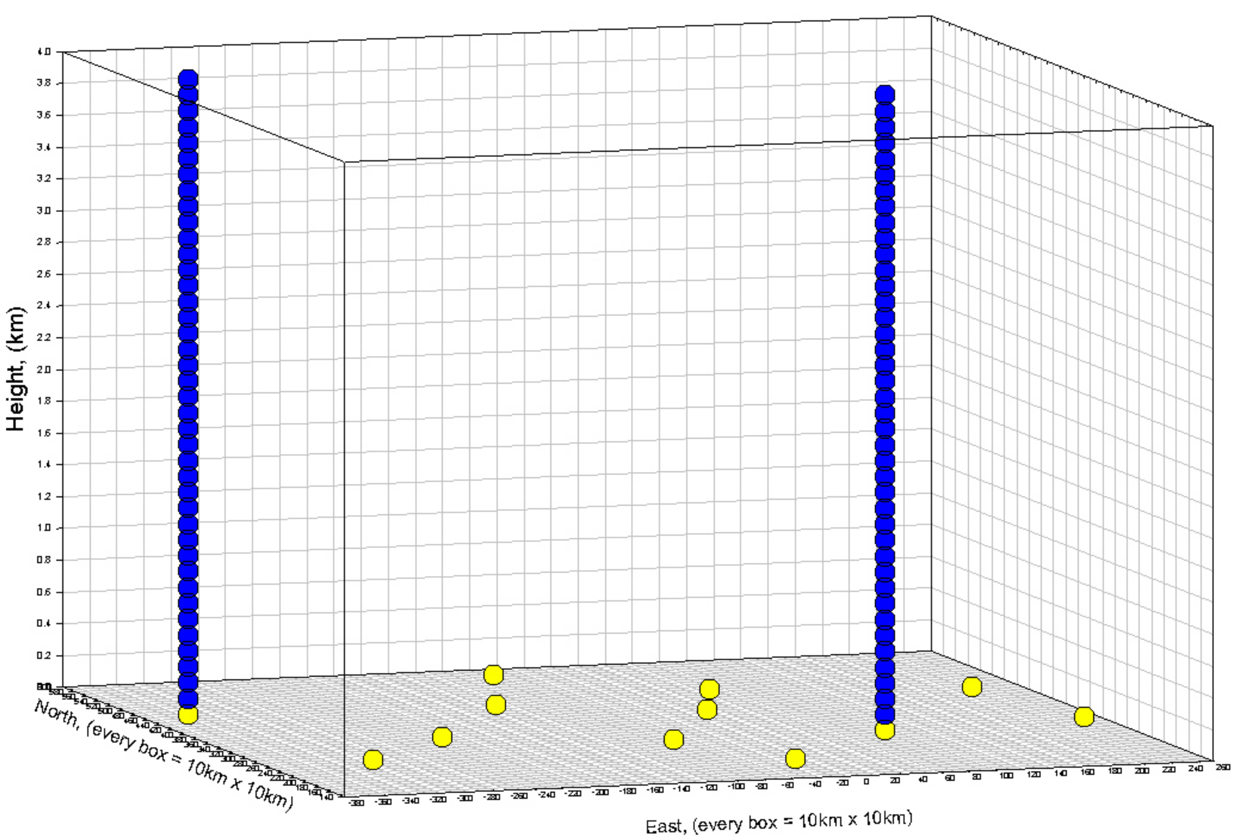
Figure 3. Horizontal Resolution and Domain of the 40-Kilometer RUC2 Model[28]



In some cases, the RUC2 data with its 40-km grid may not be of high enough resolution to capture all of the relevant flow and thermal structures that arise near the earth's surface (although the RUC2 has been available since April 2002 on a 20-km grid). To avoid this problem, some investigators (for applications such as the one described by Kaplan et al.[29]), taken advantage of a technique to introduce high-resolution terrain data and surface observations using a "mesoscale assimilation system".

In many CALPUFF applications, even those using MM5 prognostic model output with traditional airport and balloon sounding data, the area between the major sources and the receptor locations lacks significant meteorological coverage meteorological observations and requires the model to interpolate the data and fill in the grid points that have no data. The model must interpolate in space and time between the twice-daily balloon soundings, which fall near the times of sunrise and sunset in the continental United States. Due to interpolation, the model may underestimate wind speeds by missing diurnal features such as the daytime diurnal wind speed maximum or the low-level jet stream after sunset. During periods when the wind shifts nearly 180 degrees between sounding times, interpolation of vector winds can inappropriately yield near-calm winds at the midpoints of the 12-hour periods between sounding times. Even accounting for balloon sounding data, the 3-dimensional wind field is mostly devoid of real measurements, as shown in Figure 4.

Figure 4. Three-Dimensional View of Data Coverage During Sounding Periods – Example of Traditional Meteorological Data Coverage

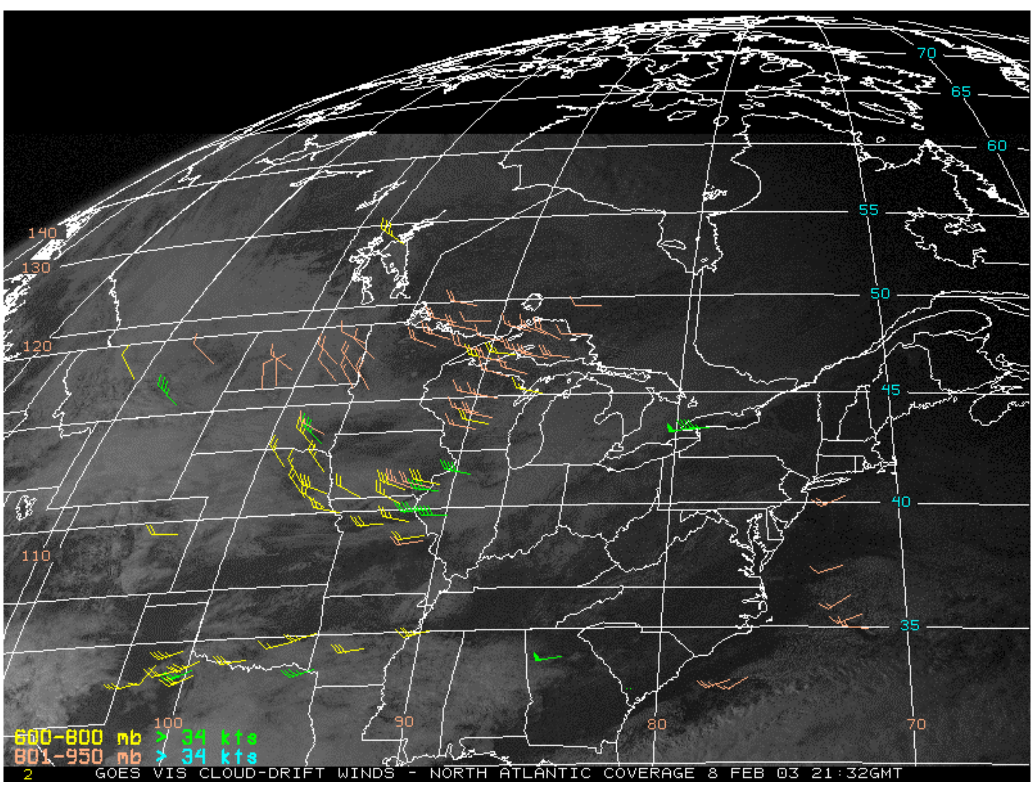


While most forecast models are initialized every three to twelve hours, the RUC model began in 1999 to be initialized every hour, making it ideal for input to dispersion models. It is a short-term weather data assimilation and forecast model that is re-initialized each hour based on the projected analysis from the previous hour and updated meteorological data readings. The major advantage of the RUC model over all other prognostic models is that it incorporates new sources of data, many of which are only available to NOAA, in addition to the hourly surface observations and twice-daily balloon soundings, such as:

- satellite derived-wind data;
- Next Generation radar (NEXRAD) that provides newly available Doppler wind data in three dimensions from several radar sweeps each hour;
- wind profilers that probe the atmosphere vertically; and
- aircraft ascent-descent reports, newly available from several hundred commercial flights per day in the U.S.

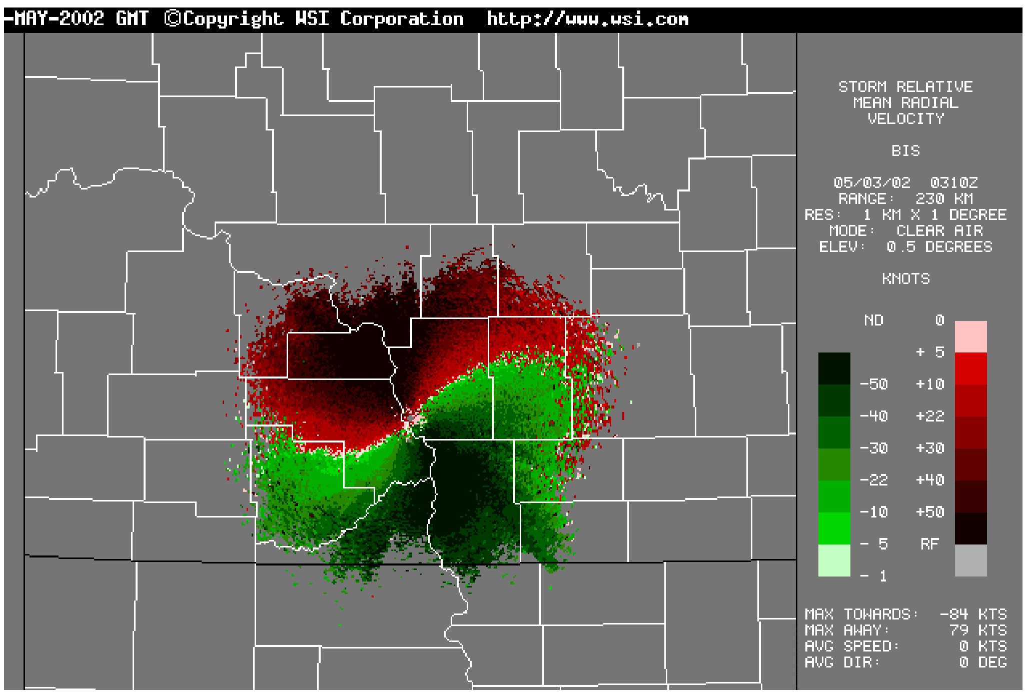
Satellites such as Geo-stationary Operational Environmental Satellite (GOES)-East and GOES-West derive wind speed and direction from cloud movement under all weather and cloud conditions over the Earth's surface using Infra-Red (IR), Water Vapor (WV), and Visible channels. Figure 5 shows one hour of wind speed and direction derived from GOES-East Visible channel.

Figure 5. GOES – East Satellite Derived Winds[30]



NEXRAD (Next Generation Radar) Doppler Radar[31] measures precipitation and wind based upon the energy and the "shift in the phase" returned to the radar when it bounces off a target. When the radar signal hits a target (i.e., raindrops, insects, or other available scattering media), the phase of the signal that returns to the radar depends on the direction that the target is moving. The radar also measures the change in phase of the object and converts it to velocity as it moves towards or away from the radar (see Figure 6). The computed velocities or VAD (Velocity Azimuthal Display) winds are incorporated into the RUC2 model. An advantage of VAD winds is the widespread coverage of NEXRAD radar across the country (Figure 7). A complete sweep of NEXRAD radar is made every 10 minutes. The availability of the NEXRAD data greatly increases the actual wind data available to the RUC2 prognostic model every hour over that of traditional data (compare Figures 8 and 4).

Figure 6. Radial Velocity Wind Components from NEXRAD Radar (Green Moving Towards the Radar, Red moving Away from the Radar)[32]



In addition to NEXRAD and satellite data, the RUC2 model incorporates aircraft ascent and descent data from over 500 flights each day (see Figure 9). Airlines such as Delta, Northwest, United, and Federal Express transmit the flight's latitude, longitude, altitude, time, temperature, wind speed and direction.

The wind profiler installations across the United States are shown in Figure 10. The profilers provide hourly soundings of wind, temperature, and turbulence data at many levels in the vertical.

The RUC2 model assimilates all available data, performs a quality assurance check, reads in the previous 1-hour RUC2 model forecast, and outputs a forecast for the next 12 hours. The data

Figure 7. Completed NEXRAD Doppler Radar Installations Within the United States[33]

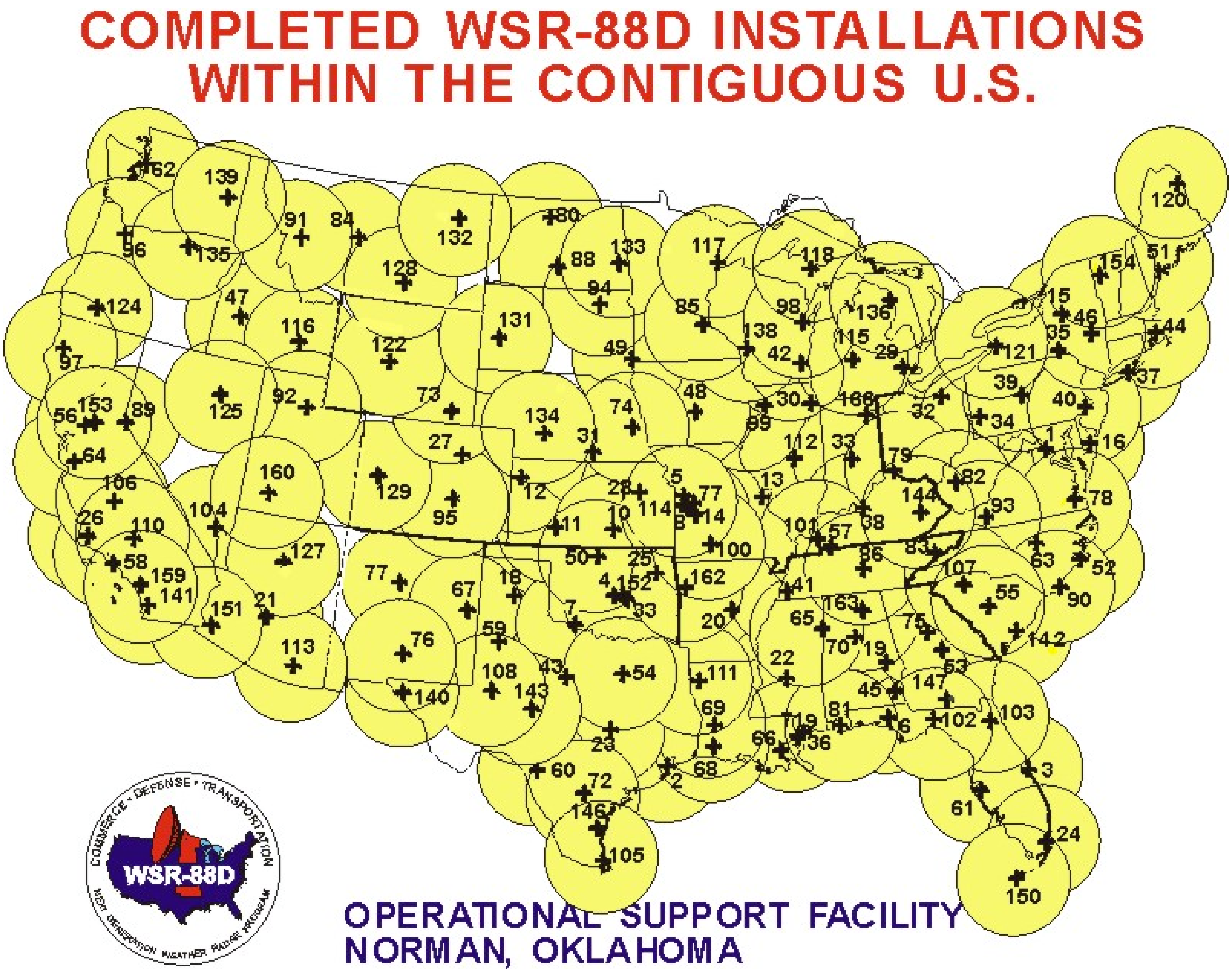


Figure 8. Example of Hourly Meteorological Data Coverage with Clear-Air NEXRAD

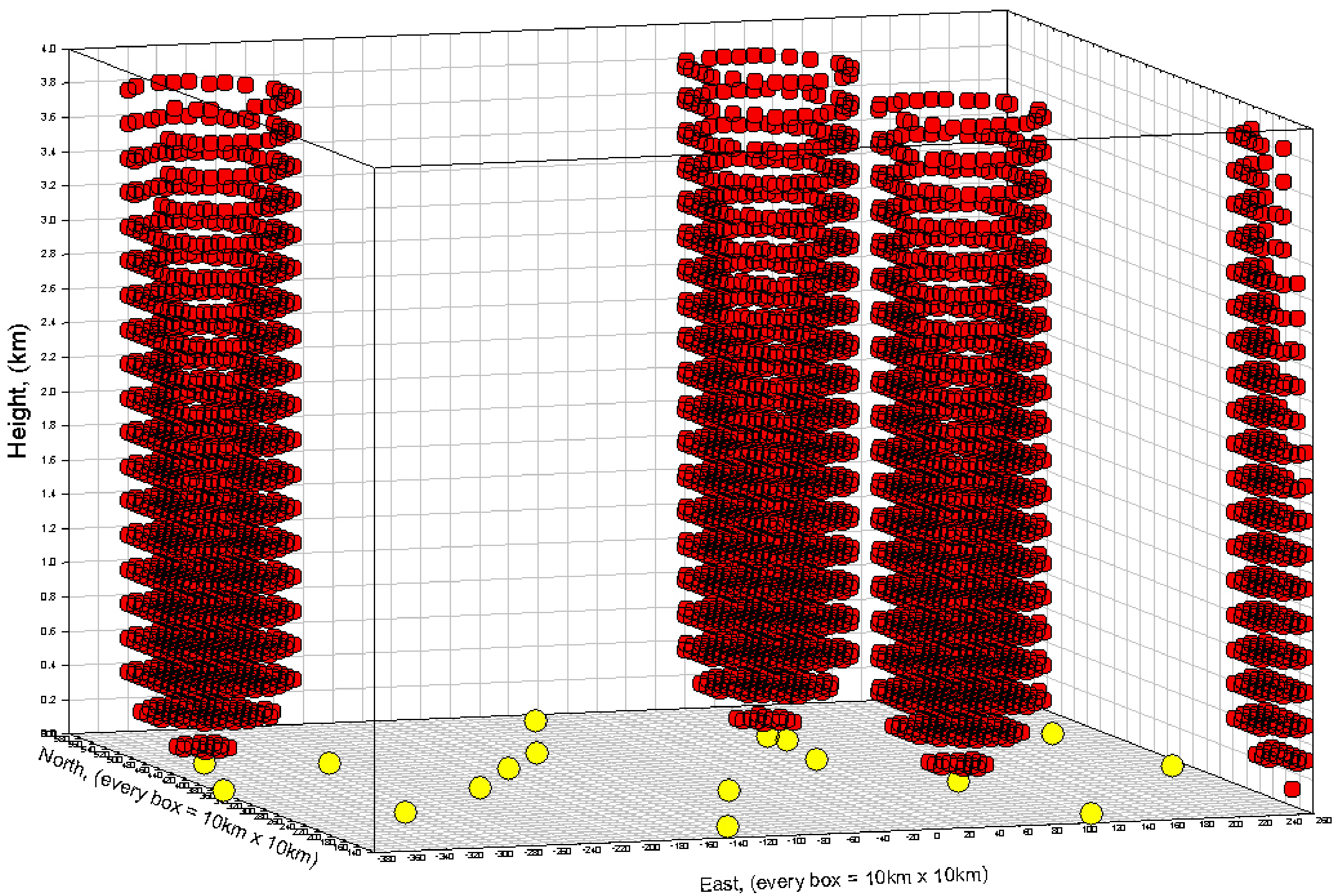


Figure 9. Typical ACARS Coverage for a 24-Hour Period Up to 5000 Feet[34]

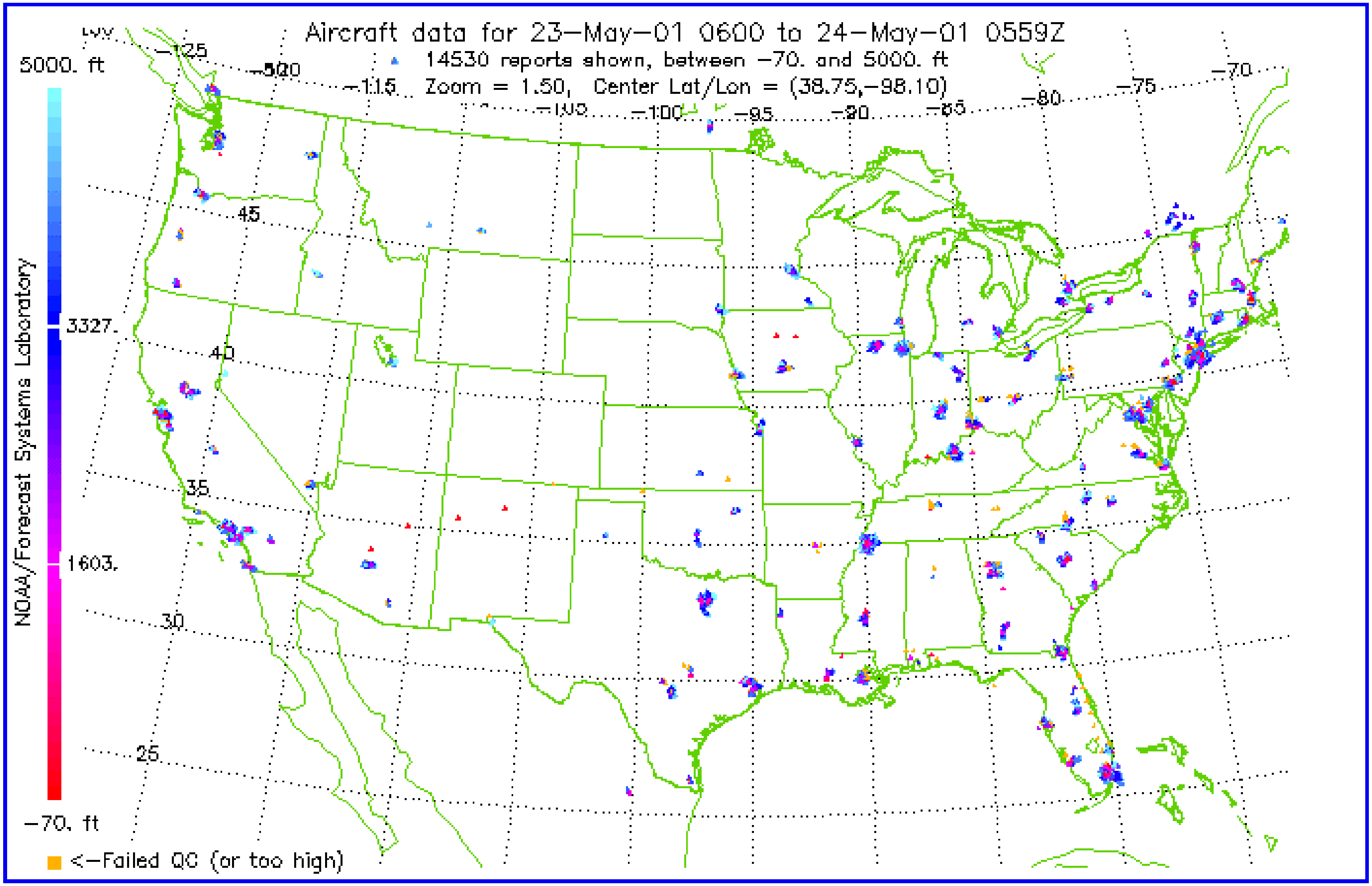
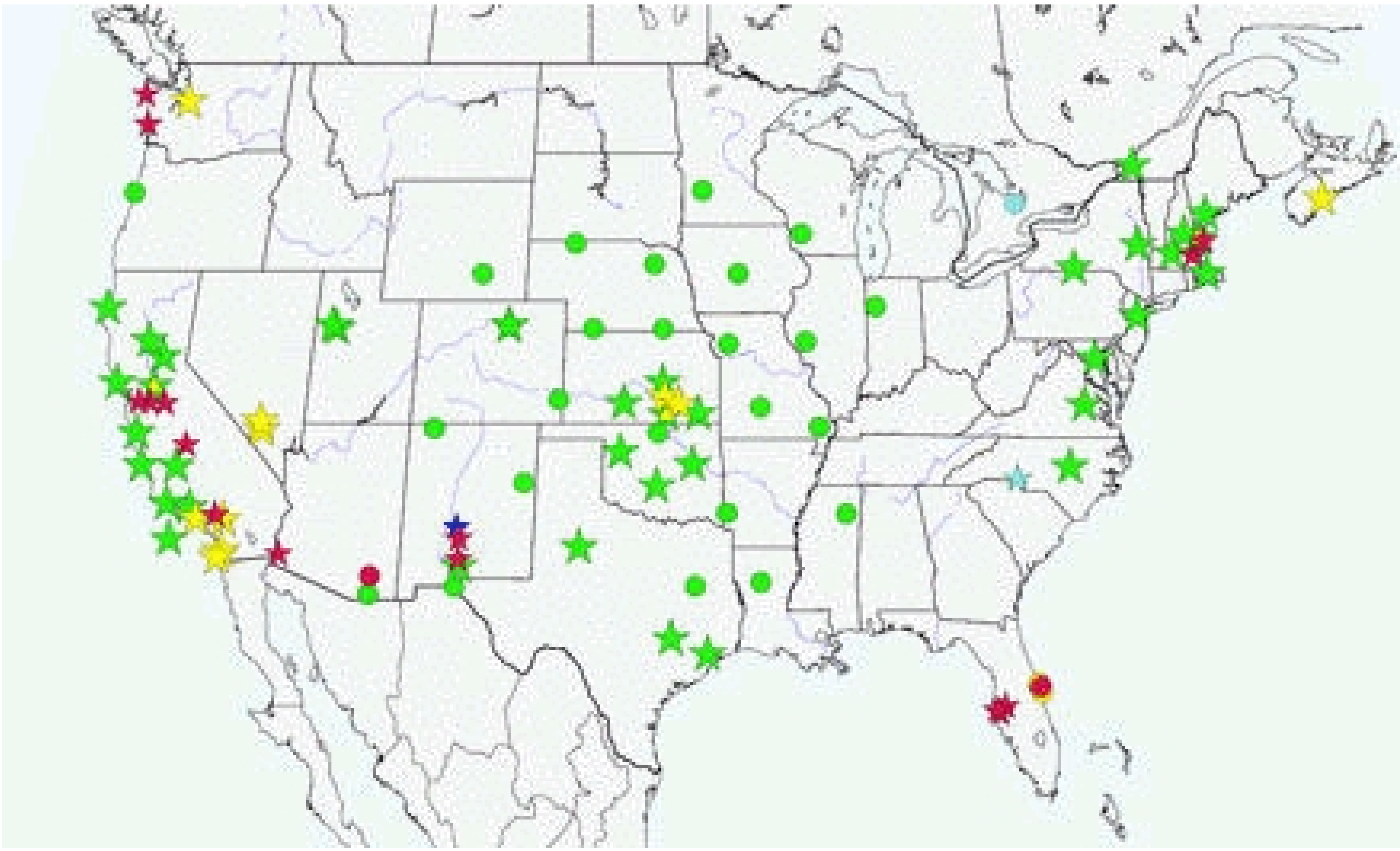


Figure 10. Coverage of Profiler Wind Data Stations in the United States



analysis and model forecast account for terrain, land/water interaction, mountain circulations, sea/lake breezes, snow cover, vegetation, soil moisture, and a host of other variables.

These new meteorological observations have the potential to increase the accuracy of CALPUFF model simulations. The use of the enhanced meteorological data, specifically the NEXRAD winds, has been found to reduce MM5 model wind errors[36], and was recommended in presentations at the EPA's Seventh Modeling Conference[37]. Kaplan et al.[29] have found in preliminary modeling runs that the RUC2 MM5 winds are sometimes higher during apparent near-stagnant conditions derived from traditional wind measurements. Comparisons made by the authors with measurements from meteorological towers throughout central and western North Dakota indicaate that the wind speeds from the RUC2 MM5 model are in better agreement than traditional CALPUFF winds derived from only balloon soundings and airport data. As a result, the CALPUFF model concentration predictions are lower than those from traditional meteorological data, while still being protective of air quality.

The use of RUC2-derived winds may also provide more realistic simulations for regional transport studies. For example, it has been found in the BRAVO tracer experiments in Texas that predicted tracer movements are slower than those actually measured[38]. Part of the reason for this lag in tracer movement could be the underestimate of winds aloft due to the paucity of real meteorological data aloft, a condition that could possibly be mitigated by the use of RUC2-derived meteorological data.

CONCLUSIONS

The adoption of the FLAG guidance and its implementation with CALPUFF has important implications for the ability for most proposed new or modified emission sources to be permitted in the United States. There are several features of the CALPUFF modeling system and the application of the FLAG procedures that add considerable and unwarranted conservatism to the results. Besides the known limitations of CALPUFF to account for plume spreading associated with nocturnal wind shear, these features include:

- omission of certain components of naturally occurring particulates, such as natural sea salt particles;
- how cloudy nighttime conditions and precipitation events can inappropriately influence the visibility assessments and should be properly accounted for; and
- how the daily averages of the ratio of the source-caused to background light extinction are calculated.

These limitations can be readily addressed through enhancements in the CALPOST software and use of IMPROVE data for sea salt concentrations. Improvements in the specification of relative humidity for regional haze impacts are possible by 1) limiting the RF in the f(RH) calculation to 95%, and 2) considering MM5 predictions of RH as "pseudo-stations" to improve the spatial resolution of RH input[39]. It is important that EPA and IWAQM adopt these enhancements as soon as possible to eliminate the excessive conservatism and associated lack of scientific credibility associated with the current FLAG procedures.

The availability of enhanced meteorological data for use in CALPUFF applications from the RUC NOAA model is an important new development. The use of traditional meteorological observations from airports and balloon soundings creates large holes in the four-dimensional wind field that is used in CALPUFF assessments. These data gaps have the potential of creating significant errors in the winds (especially wind speed underestimates), most apparent when the winds shift between sounding times. Initial testing of RUC2 data by Kaplan et al.[29] and comparisons of these winds with tower data in North Dakota provide encouraging results for this type of meteorological data in long-range transport modeling applications.

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KEYWORDS

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Visibility assessment

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FLAG

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Regional haze

Prognostic Meteorological Model