

October 21, 2005

Ms. Rosalina Rodriguez  
Planning Section, Central Office  
Division of Air Quality  
North Carolina Department of Environment and Natural Resources

Re: Comments on VISTAS's Draft Regional Haze Modeling Protocol

Dear Ms. Rodriguez:

The National Lime Association (NLA) appreciates the opportunity to comment on VISTAS's draft regional haze modeling protocol and BART control technology document. NLA is the trade association for manufacturers of high calcium quicklime, dolomitic quicklime, and hydrated lime, collectively referred to as "lime." Lime provides cost-effective solutions to many of society's manufacturing and environmental needs.

The lime manufacturing process involves combustion of fossil fuels to calcine limestone in kilns. Kiln emissions include NO<sub>x</sub>, SO<sub>2</sub>, and particulate matter. Fifteen BART-eligible lime kilns are included in VISTAS's list of BART-eligible sources. These kilns collectively contribute less than 1/10<sup>th</sup> of 1 percent of regional haze-related emissions in the VISTAS Region (Table 1).

NLA's comments address four issues:

1. For screening simulations to be useful, reasonable criteria must be used. By contrast, VISTAS's draft protocol uses unduly conservative criteria (e.g., worst-case estimate from a 3-year simulation, based on very conservative modeling).
2. AP-42 emissions factors for condensable PM for the lime industry are rated "poor" by EPA. The AP-42 estimates are indeed "poor" and should not be used by the states to "gap-fill" missing data.
3. Lime companies that will be upgrading their air pollution control devices to comply with the lime MACT should be allowed to use projected 2007 PM emissions (i.e., the MACT PM limits), rather than 2002 PM data.
4. The VISTAS control technology document incorrectly assumes that control technologies feasible for cement kilns are transferable to lime kilns.

#### **Screening Modeling Should be Based on Moderately Conservative Criteria**

VISTAS's draft BART screening assessment employs modeling assumptions intended to ensure that sources that contribute to visibility impairment are not exempted in error (Draft Protocol, at pg. 31). VISTAS has characterized the methodology as using "moderate conservatism [because]

screening procedures that are so conservative that few sources will pass the screen are in practice not very useful” (Id. at 33). Given that there are 275 BART-eligible sources in VISTAS, and SIP revisions are due to EPA in 14 months, it is essential that VISTAS’s modeling assumptions reflect reasonable worst case assumptions -- if for no other reason, to avoid state agencies having to devote scarce resources reviewing or performing screening analyses that have virtually no chance of excluding any sources.

One assumption which stands out as not meeting VISTAS “moderate” conservatism standard is the requirement that the *highest* impact value over a three-year simulation period be used in making a determination of no contribution to visibility impairment. By contrast, EPA uses the 98<sup>th</sup> percentile of modeled visibility values, explaining that such an approach

exclude[s] roughly 7 days per year from consideration ... effectively captur[ing] the sources that contribute to visibility impairment in a Class I area, while minimizing the likelihood that the highest modeled visibility impacts might be caused by unusual meteorology or conservative assumptions in the model (70 Fed. Reg. 39,121).

NLA urges VISTAS to use the 98<sup>th</sup> percentile (8<sup>th</sup> highest annual), rather the highest predicted impact value. By so doing, VISTAS will achieve its goal of employing a screening approach that imparts some practical value.

### **States Should Not “Gap-Fill” Missing Data for Condensable PM Emissions Based on AP-42 Emission Factors**

VISTAS’s draft BART protocol states that the following emissions information (listed in order of priority) should be used for BART modeling:

- 24 hour maximum value emissions for the period 2001-03 (CEM data)
- 24 hour maximum value for 2002 (CEM data)
- Actual 2002 emissions in state CERR inventory
- AP-42 source profiles
- Permit allowable emissions
- Potential to emit

Regarding PM<sub>2.5</sub>, continuous emission monitors for condensable emissions do not exist. Nor are test data available for most lime kilns in the VISTAS Regions. Lime plants have refrained from using Method 202 due to its well-recognized, significant positive biases (see Attachment A).

Although AP-42 emission factors (EF) have been published for lime kilns, EPA has assigned a quality rating of “E” (i.e., poor) to these EFs. For preheater kilns, the EF is based on a single test of a straight (not a preheater) kiln. The EF is highly suspect because it is nearly 3 times the EF for straight rotary kilns. Due to the greater inherent scrubbing of gases with lime in preheater kilns, such kilns would be expected to emit less (not more) condensable PM. For straight kilns, EPA has specified an EF of 0.38 lb/ton lime – yet, this value is three times the median value of the tests used to derive the EF. Attachment B provides additional details on why the AP-42 EFs are flawed.

For lime kilns, condensable PM emissions result primarily from the condensation of sulfur and/or nitrogen compounds. A portion of these “condensable particulates” have already been accounted for in the CERR as part of the monitoring of their gaseous counterparts, which remained gases at the stack temperature where they were being monitored.

Given the substantial limitations in measuring condensable emissions as described in Attachment A, and the potential for double-counting these emissions (i.e., in their gaseous and condensable forms), NLA recommends that condensable PM<sub>2.5</sub> not be included in BART modeling for lime kilns until a more refined and useable test method is developed. In addition, because the EFs in AP-42 are of poor quality, states should not “gap-fill” emission estimates based on these EFs.

### **Lime Plants Should be Allowed to Model PM<sub>10</sub> Based on PM MACT Limits**

Most BART-eligible like kilns in the VISTAS Region will be subject to the Lime MACT. For several of these kilns, pollution control upgrades will need to be installed to comply with the Lime MACT before Jan. 5, 2007. These upgrades will decrease PM emissions below 2002 levels. For such kilns, the permissible sources of emissions data listed above should be expanded to allow the use of projected 2007 PM emissions.

### **Control Technologies for Cement Kilns are Not Transferable to Lime Kilns**

#### **NO<sub>x</sub> Controls**

VISTAS has identified the following NO<sub>x</sub> control technologies (and related costs) for lime manufacturing plants:

<b>Control</b>	<b>Control Efficiency (%)</b>	<b>Cost/ Ton Reduced</b>
Mid-Kiln Firing	30	\$460
LNB	30	\$560
SNCR – Urea Based	50	\$770
SNCR - NH <sub>3</sub> Based	50	\$850
SCR	80	\$3,370

This information is derived from EPA’s AirControlNET database, which in turn references EPA’s 1994 Alternative Control Technologies document for cement kilns.<sup>1</sup> However, lime kilns and cement kilns are very different. Lime kilns are used to calcine chemical grade limestone into high purity lime. By contrast, cement kilns process a mix of materials to create clinker. Requirements for purity, and combustion practices to ensure that purity, are dramatically different between the two industries. In addition, the raw feedstock for cement kilns differs substantially from feedstock for lime kilns. Whereas the particle size of feed to a cement kiln is measured in microns (e.g., 80% less than 75 microns), limestone fed to a lime kiln is measured in inches (e.g., 1 inch to 2½ inches). This difference in feed particle size means that some technologies possible on cement kilns are not feasible on lime kilns.

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<sup>1</sup> USEPA, Office of Air Quality Planning and Standards. *Alternative Control Technologies Document – NO<sub>x</sub> Emissions from Cement Manufacturing*. EPA-453/R-94-004 [hereinafter referred to as Cement NO<sub>x</sub> ACT Document]

In the 1998 NO<sub>x</sub> OTAG Transport Rulemaking, EPA examined the appropriateness of imposing the control technologies listed above on the lime industry. According to EPA estimates, in 1997, lime plants emitted 2/10<sup>th</sup>s of 1 percent of NO<sub>x</sub> in the OTAG Region. Significantly, in the 1998 Rule, EPA recommended to the OTAG states that lime plants not be targeted for NO<sub>x</sub> reductions (63 Fed. Reg. 56,416). This contrasts with the cement industry, for which EPA recommended that 30% NO<sub>x</sub> reductions should be sought because control technologies were available that fell within EPA's \$2000/ton of NO<sub>x</sub> removed cost-effectiveness range (*Id.* at 57,418).

The substantial disparity in the production rates of cement vs. lime kilns must be taken into account when evaluating the affordability of control devices. The average capacity of cement kilns is about 1650 ton/day, whereas the average capacity of BART-eligible lime kilns in the VISTAS Region is 600 ton/day. EPA and the states have generally recognized a 0.6 power (i.e., exponent) factor to be appropriate when scaling control equipment costs. At a minimum, control costs for cement kilns would need to be scaled by this factor to adjust for any technologies deemed to be technically feasible.<sup>2</sup> Furthermore, cost estimates based on new construction should be adjusted to take into account that retrofit costs would typically be much higher. In addition, it may be impossible for some plants to retrofit their equipment due to the physical layout of their operations.

Since the NO<sub>x</sub> Rule was promulgated in 1998, PSD permits for 11 lime kilns have been issued. For each kiln, EPA has agreed with the states' determinations that the post-combustion NO<sub>x</sub> controls listed above are either technically infeasible or cost prohibitive for lime kilns. Instead, permits for these kilns define BACT as efficient combustion practices, minimization of fuel consumption and excess air for the combustion process, or some similar control techniques (see EPA Clearinghouse). The average capacity of these 11 new kilns is about 1100 tons/day, nearly twice the 600 tons/day capacity of the median BART-eligible lime kiln in the VISTAS region. Because NO<sub>x</sub> control costs would be spread over fewer tons of lime for the 15 BART-eligible kilns, the cost-effectiveness of NO<sub>x</sub> controls for them would be less "reasonable" than for the 11 kilns subject to BACT review (see 0.6 power factor comment above).

Set forth below is a discussion of the feasibility of the NO<sub>x</sub> control technologies identified by VISTAS.

#### Mid-Kiln Firing (MKF)

Mid-kiln firing is a form of staged combustion of fuels. A specially designed fuel injection system introduces a second fuel source at a midpoint in the kiln. This system is typically used in cement kilns and allows the fuel to be burned at a temperature of 600-900°C (1100-1650°F), which is much lower than the burning temperature of 1950-2050°F in a lime kiln.<sup>3</sup>

By adding fuel at mid-kiln, MKF changes the flame temperature and length. These changes may reduce thermal NO<sub>x</sub> formation in a cement kiln by burning part of the fuel at a lower

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<sup>2</sup> Cement NO<sub>x</sub> ACT Document, pg. 6-24. When applying a 0.6 power factor to costs/ton of production based on a 1650 tpd kiln, costs/ton of production for a 600 tpd kiln are 50 percent higher.

<sup>3</sup> Battye et al., EC/R Incorporated, "NO<sub>x</sub> Control Technology for the Cement Industry." Final report prepared for USEPA, September 19, 2000, page 65.

temperature and create reducing conditions at the primary fuel injection point which may destroy some of the NO<sub>x</sub> formed upstream in the kiln burning zone. The discontinuous MKF feed that results from introduction of staged fuels at the kiln midpoint (i.e. introduction of a solid fuel once every revolution) can result in less stable combustion conditions and increased CO emissions depending on the type of fuel used.

Although MKF may reduce NO<sub>x</sub> emissions in cement kilns, it is technically infeasible for lime kilns. The addition of fuel to the lime at mid kiln will have a negative effect on the quality of the lime produced. Introduction of fuel at mid-kiln will increase carry-over of unburned carbon to the product. This unburned fuel will prevent the lime product from being used in many applications.

### Low NO<sub>x</sub> Burners (LNB)

Although some burners are marketed specifically as LNBs, there is no specific definition of what qualifies a burner as an LNB. The U.S. EPA's *Alternative Control Technology Document NO<sub>x</sub> Emissions from Cement Manufacturing* indicates that burners specifically marketed as LNBs "typically use 5 to 7 percent primary air." Reduced primary air is one characteristic of a LNB.

EPA has indicated that a 20 to 30 percent reduction in NO<sub>x</sub> emissions may be anticipated in cement kilns by switching from a direct-fired standard burner to an indirect-fired LNB. However, EPA has determined that "the [emission reduction] contribution of the low-NO<sub>x</sub> burner itself and of the firing system conversion [from direct to indirect] cannot be isolated from the limited data available."<sup>4</sup>

In lime manufacturing, direct and indirect firing describe the manner in which pulverized fuel is conveyed from the fuel-grinding mill to the burner.

Direct Firing Systems. In the direct firing configuration, fuel is pneumatically conveyed directly from the coal mill to the burner. The quantity of air introduced to the primary combustion zone is dictated by the minimum sweep air requirements of the coal mill and the conveyance system rather than by the optimum flame requirements. All BART-eligible kilns in the VISTAS region use a direct firing system.

Indirect Firing Systems. In the indirect firing system, the coal is pulverized in the coal mill and pneumatically transported to a storage tank. The pulverized fuel is then conveyed to the burner with the quantity of air that is optimum for flame considerations. This combustion air is completely independent of the sweep air requirements of the coal milling system. There have been no controlled studies conducted on lime kilns that verify that this method of burning solid fuels reduces the formation of NO<sub>x</sub>.

The cost of converting existing firing systems on BART-eligible lime kilns in the VISTAS Region to indirect firing would be cost-prohibitive, and certainly well above the \$560/ton of NO<sub>x</sub> removed in VISTAS's control options document.<sup>5</sup> Such a conversion would require,

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<sup>4</sup> Cement NO<sub>x</sub> ACT Document, page 5-35

<sup>5</sup> The reference for the \$560 estimate is EPA's AirControlNET database, which references EPA's 1994 Cement NO<sub>x</sub> ACT Document. The ACT document, however, does not estimate \$560 per ton. Rather, it estimates that

among other things, replacing the entire fuel handling system (coal mill, silos, etc), collectively costing several million dollars.

The above discussion pertains to rotary kilns. Vertical kilns have flameless combustion, thus rendering LNB non-applicable.

### Selective Non-Catalytic Reduction (SNCR)

This technology has been determined to be technically or economically infeasible in all BACT determinations for lime kilns. A common theme in the RACT/BACT analyses is that SNCR is infeasible because it requires a high but very specific temperature range (between 1600-2100°F) to be effective. At lower temperatures, the NO<sub>x</sub> reduction reaction is incomplete, and at higher temperatures NO<sub>x</sub> emissions can actually be increased.

There are two possible locations where the reagent (urea or ammonia) could be injected where this temperature profile could be achieved: after the air pollution control device (in most cases a baghouse), or within the kiln. Regarding the first option, flue gas exhaust temperatures from lime kilns (generally about 450°F) are substantially below the SNCR operating range. Consequently, the exhaust gases would need to be reheated. Recent BACT analyses estimate that the cost of reheating alone -- excluding reagent cost -- would result in an average cost/ ton of NO<sub>x</sub> removed greater than \$8,000.<sup>6</sup> If more current natural gas prices were assumed, this cost would exceed \$15,000/ton of NO<sub>x</sub> removed. Reheating exhaust gases would also result in additional pollutants being formed from the combustion products from the fuel used to reheat the exhaust gases.

Turning to the second option, several RACT/BACT analyses address the infeasibility of injecting the reagent within the kiln. For a fluidized bed kiln, this would require installation of nozzles and flow straighteners, which would not only be inappropriate for this type of kiln, but could also adversely affect the process chemistry. So too, with a vertical kiln, injection of the reagent within the kiln would contaminate the product. For a straight rotary kiln, temperatures in the reagent injection region would result in increased NO<sub>x</sub> formation. For a preheater kiln, the regions where optimum temperatures exist in a lime kiln contain either large stone or have exceedingly short residence times. Large stone would either damage spray nozzles, or the sprays would impinge on the stone, wetting the stone but not entraining the reagent in the gas stream.

Another concern raised in BACT analyses with using SNCR is the formation of unreacted ammonia or urea that will react with sulfur oxides in the flue gas in the presence of water to form ammonium bisulfite (NH<sub>4</sub>HSO<sub>4</sub>), a sticky compound that can cause corrosion, fouling, and blockages downstream of the injection point. This would create serious problems with the preheater, ductwork, baghouses, and fans; reduce kiln draft; and cause excessive outages. Furthermore, ammonia slip from such systems would increase formation of secondary fine particulate in the atmosphere. Finally, ammonia absorption into the lime kiln dust collected in the baghouse would seriously impact, if not eliminate the ability to sell this byproduct. Not only

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retrofitting LNBs in existing direct-fired long dry and preheater cement kilns costs from \$970 to \$1,330 per ton (Tables 2-5 and 6-17).

<sup>6</sup> In the PSD permit analysis for 625 tpd kiln at U.S. Lime & Mineral's Arkansas Plant, exhaust gas reheat is estimated to be \$8,322/ton NO<sub>x</sub> removed, at \$6.45/MMBtu natural gas.

maintaining but also increasing the sale of these byproducts is a key industry strategy in meeting our CO<sub>2</sub> intensity reductions under DOE's Climate Vision Program.

For all of the above reasons, SNCR is not a feasible NO<sub>x</sub> control strategy for the lime industry.

### Selective Catalytic Reduction (SCR)

Selective catalytic reduction (SCR) is an exhaust gas treatment process in which ammonia (NH<sub>3</sub>) is injected into the exhaust gas upstream of a catalyst bed. On the catalyst surface, NH<sub>3</sub> and nitric oxide (NO) or nitrogen dioxide (NO<sub>2</sub>) react to form nitrogen and water

To achieve the 80% removal rate listed in VISTAS's control document, an SCR would need to be operated at a temperature between 700 and 750°F.<sup>7</sup> Efficient operation of an SCR process requires fairly constant exhaust temperatures (usually ± 200°F).<sup>8</sup> Fluctuation in exhaust gas temperatures reduces removal efficiency. If the temperature is too low, ammonia slip occurs. Ammonia slip is caused by low reaction rates and results in both higher NO<sub>x</sub> emissions and appreciable ammonia emissions. If the temperature is too high, oxidation of the NH<sub>3</sub> to NO can occur. Other emissions potentially generated by SCR include increased PM emissions (from ammonia salts in a detached plume) and increased SO<sub>3</sub> emissions (from oxidation of SO<sub>2</sub> on the catalyst).

To avoid fouling the catalyst bed with the PM in the exhaust stream, an SCR unit would need to be located downstream of the particulate matter control device. However, due to the low exhaust gas temperature exiting air pollution control devices at lime plants (in most cases, a baghouse), a heat exchanger system would be required to reheat the exhaust stream to the desired reaction temperature. The cost of exhaust gas reheat would be prohibitive, as with the SNCR systems noted above.

Although SCR is being used in the utility industry, there are significant differences between the exhaust streams generated by the two industries that account for the difference in the application of the technology. A utility boiler's exhaust gas stream does not vary over time. That is, the gas stream characteristics do not change greatly, whereas, the lime kiln exhaust gas stream temperature has a high degree of fluctuation. The temperature variability of the exhaust gas stream makes the technology technically infeasible.

### **SO<sub>2</sub> Controls**

Control	Control Efficiency (%)	Cost/ Ton Reduced
FGD	90	\$0

The VISTAS control technology summary suggests that 90% SO<sub>2</sub> removal can be achieved by "FGD" control, at no cost. A VISTAS representative recently informed NLA that the 90% removal rate is based on the inherent removal of SO<sub>2</sub> in lime kilns. Conversely, the background

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<sup>7</sup>Air Pollution Control Cost Manual, Sixth Edition, January 2002, Section 4.1, Chapter 1, Selective Catalytic Reduction, NO<sub>x</sub> Controls, EPA/452/B-02-001, page 2-10.

<sup>8</sup>Id, page 2-11.

document from AirControlNET indicates that the 90% removal is based on add-on wet, dry, and semi-dry FGD systems.

Of the 15 BART-eligible lime kilns in VISTAS, 6 are preheater kilns, for which a 90% inherent removal is a reasonable estimate (depending on the residence time of the stone, and a host of other factors). For straight rotary kilns (as well as single shaft vertical kilns), however, a 90% removal rate based on inherent scrubbing within the kiln is less likely than for preheater kilns, again depending on kiln-specific factors.

Turning to the feasibility of scrubbers, 10 of the 11 PSD permits issued to lime kilns since 1998 are for preheater kilns, with inherent scrubbing within the kiln deemed to be BACT. Only one permit has been issued for a straight rotary kiln, a 1050 tpd kiln at Graymont Inc.'s plant in Pennsylvania. A wet scrubber was required for the kiln because the estimated cost/ton of SO<sub>2</sub> removed was less than \$5,000 -- PADEP's affordability threshold for BACT for SO<sub>2</sub>. This high cost threshold would not be warranted for existing BART sources where such technology would be applied as retrofit. Graymont's straight rotary kiln has not been built, and thus it is unknown whether the required removal rate will be achieved. Space constraints for a scrubber will also be a factor for some of the BART-eligible kilns at lime plants in the VISTAS Region.

### **PM Controls**

For lime kilns, the VISTAS control technology document indicates that 99% PM control efficiency is feasible with fabric filters and ESPs. It should be noted that wet scrubbers are also capable of achieving these PM removal rates. Particulate matter from BART-eligible lime kilns contribute only 1/100<sup>th</sup> of 1 percent of regional haze-related emissions in the VISTAS Region (Table 1).

In addition to lime kilns, VISTAS identifies various control technologies for several other PM sources at lime plants, including primary crushing, secondary crushing/screening, raw material transfer and conveying, raw material unloading, hydrators, raw material storage piles, lime silos, packing/shipping, product transfer and conveying, and product loading.

Regarding the above units that process limestone, most PM emitted from these units is not PM<sub>2.5</sub>, but rather is coarse material larger than PM<sub>10</sub>. This material does not contribute to PM<sub>2.5</sub> nonattainment, nor does it contribute to regional haze. Consequently, such units should not be subject to BART reviews.

Furthermore for all non-kiln units, it is important for the states to limit RACT reviews to those units installed between 1962 and 1977. Because components in material handling systems at lime plants are regularly replaced due to wear, only a fraction of the emissions shown in VISTAS' inventory are potentially subject to the BART rule. Based on the fact that material handling sources do not emit PM<sub>2.5</sub> precursors (SO<sub>2</sub> and NO<sub>x</sub>) and that PM emitted from such sources are large particles (i.e., greater than PM<sub>10</sub>), NLA suggests that VISTAS exempt all non-kiln units at lime plants from further consideration under this rule.

Conclusion

BART-eligible sources at lime plants account for less than 1/10<sup>th</sup> of 1 percent of regional haze-related emissions in the VISTAS Region. Yet, by number, they constitute 5 percent of the 275 sources for which VISTAS has indicated BART modeling will have to be conducted. NLA recommends that VISTAS forego subjecting lime plants to its BART modeling program. Particulate matter will be substantially reduced from these sources due to the Lime MACT, whose effective date pre-dates the Regional Haze Rule. For the other two haze-generating pollutants (NO<sub>x</sub> and SO<sub>x</sub>), ten BACT analyses that have been subject to state, US EPA and public review, have concluded that affordable control technologies are unavailable to reduce these pollutants for kilns comparable to those in the VISTAS Region. Given these facts, most other RPOs have decided not to target lime plants for haze-related reductions. This decision to focus administrative resources on source categories with greater emissions is fully consistent with EPA's advice to states in the 1998 OTAG Transport Rulemaking SIP (63 Fed. Reg. 57,416). As noted above, in 1997, EPA estimated that lime plants emitted 2/10<sup>th</sup> of 1 percent of NO<sub>x</sub> in the OTAG Region.

If you have any questions regarding these comments, please do not hesitate to contact me at (703) 243-5488.

Sincerely,



Arline M. Seeger  
Executive Director

cc: Leigh Barb Bacon, ADEM  
Jeff Kitchens, ADEM  
Mike Kiss, VDEQ  
Larry Sorrels, EPA

**TABLE 1: Emissions from Lime Plants in VISTAS Region**

Pollutant	Company	Plant	Kiln #	Actual Emissions (tpy, 2002 NEI)		BART-Eligible (B-E) Emissions from Lime Plants as a Percent of Emissions from VISTAS Sources
				BART-Eligible Kilns	Plant Non-Kiln**	
<b>NOx</b>				<b>3,631</b>	<b>1</b>	<b>0.07 (B-E kilns)</b>
	Carmeuse	Black River	1	272	0	
	Carmeuse	Black River	2	inactive		
	Carmeuse	Black River	3	588		
	Carmeuse	Longview	2	95	0	
	Carmeuse	Longview	3	197		
	Carmeuse	Maysville	1	374	1	
	Carmeuse	Maysville	2	354		
	Carmeuse	Maysville	3	366		
	Chemical Lime	Alabaster	1	115	0	
	Chemical Lime	Alabaster	2	103		
	Chemical Lime	Kimballton	3	274	0	
	Chemical Lime	Montevallo	3	339	0	
	Chemical Lime	Montevallo	4	275		
	O-N Minerals	Chemstone	1	256	0	
	O-N Minerals	Chemstone	2	24		
<b>SO<sub>2</sub></b>				<b>2,546</b>	<b>0</b>	<b>0.05 (B-E kilns)</b>
	Carmeuse	Black River	1	21	0	
	Carmeuse	Black River	2	inactive		
	Carmeuse	Black River	3	50		
	Carmeuse	Longview	2	38	0	
	Carmeuse	Longview	3	47		
	Carmeuse	Maysville	1	40	0	
	Carmeuse	Maysville	2	33		
	Carmeuse	Maysville	3	40		
	Chemical Lime	Alabaster	1	105	0	
	Chemical Lime	Alabaster	2	94		
	Chemical Lime	Kimballton	3	639	0	
	Chemical Lime	Montevallo	3	733	0	
	Chemical Lime	Montevallo	4	704		
	O-N Minerals	Chemstone	1	2	0	
	O-N Minerals	Chemstone	2	0.2		

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**TABLE 1: Emissions from Lime Plants in VISTAS Region, continued**

Pollutant	Company	Plant	Kiln #	Actual Emissions (tpy, 2002 NEI)*		BART-Eligible (B-E) Emissions from Lime Plants as a Percent of Emissions from All VISTAS Sources
				BART-Eligible Kilns	Plant Non-Kiln**	
<b>PM-10*</b>				<b>187*</b>	<b>319**</b>	<b>0.01 (B-E kilns)</b>
						<b>0.02 (non-kiln)</b>
	Carmeuse	Black River*	1	4	43	
	Carmeuse	Black River*	2	inactive		
	Carmeuse	Black River*	3	9		
	Carmeuse	Longview	2	10	0.4	
	Carmeuse	Longview	3	10		
	Carmeuse	Maysville*	1	9	156	
	Carmeuse	Maysville*	2	8		
	Carmeuse	Maysville*	3	9		
	Chemical Lime	Alabaster*	1	13	21	
	Chemical Lime	Alabaster*	2	20		
	Chemical Lime	Kimballton	3	25	6	
	Chemical Lime	Montevallo*	3	15	86	
	Chemical Lime	Montevallo*	4	12		
	O-N Minerals	Chemstone	1	15	8	
	O-N Minerals	Chemstone	2	30		

\* PM-10 PRI reported when available. Plants with asterisk have only PM-PRI available. Thus these totals are an overestimate of PM-10.

\*\* These totals significantly overstate emissions from BART-eligible non-kiln sources because all non-kiln sources at the plant are included, not just those constructed in 1962-1977.

**ATTACHMENT A:**

**Biases Associated with U.S. EPA Reference Method 202 for the  
Measurement of Condensable Particulate Matter Emissions**

Report to:

National Lime Association  
Arlington, VA

Report Prepared by:

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August 2005

# **Biases Associated with U.S. EPA Reference Method 202 for the Measurement of Condensable Particulate Matter Emissions**

This report summarizes the technical information available concerning the significant biases associated with the use of U.S. EPA (“EPA”) Reference Method 202 for the measurement of condensable particulate matter emissions from a wide variety of stationary combustion sources. Technical information compiled since Method 202 was promulgated by EPA in 1991 clearly indicates that this method is subject to significant positive biases that are difficult to quantify and can vary significantly over short time periods at specific sources.

## **1. Summary**

Concerns regarding condensable particulate matter emissions first received EPA’s attention in the late 1970s and have recently increased dramatically as EPA prepares to implement National Ambient Air Quality Standards (“NAAQS”) for PM<sub>2.5</sub> particulate matter. Condensable particulate matter is suspected by EPA to be a major component of ambient PM<sub>2.5</sub> particulate matter.

Many Maximum Achievable Control Technology (“MACT”) regulations and Standards of Performance for New Sources (“NSPS”) regulations have been limited to filterable particulate matter emissions. These emissions are defined as particulate matter (“PM”) that exists at a temperature set by the air emission testing procedures, usually 248°F ± 25°F. Vapor phase material present in the sample gas streams during measurement of filterable particulate matter passes through the sampling train filters and is not collected. EPA has argued that the condensable vapors not collected in a filterable particulate matter stack testing procedure can form particles once the stack emissions cool in the atmosphere. Accordingly, EPA has been strongly encouraging state and local agencies to request condensable particulate matter emission tests along with filterable particulate matter emission tests.

EPA has encouraged the use of combined sampling trains to simultaneously measure filterable and condensable particulate matter emissions. Total particulate matter emissions can be measured by the combination of a Method 5 (filterable PM) and a Method 202 (condensable PM) sampling train. PM<sub>10</sub> particulate matter can be measured by the combination of a Method 201A (filterable PM<sub>10</sub>) and a Method 202 (condensable PM) sampling train. Both Method 5 and Method 201A include a set of water-filled, ice cooled impingers after the filter for sample gas moisture measurement. When condensable particulate matter data are also needed, Method 202 provides procedures for the recovery of inorganic and organic material captured in these impingers.

Significant measurement errors are associated with EPA’s Method 202. These errors are due, at least in part, to the absorption and subsequent oxidation of inorganic gases and the absorption of soluble gaseous organic compounds to form condensable material in the Method 202 impinger solutions. Gases are not defined as condensable vapors and should not be captured in the condensable particulate matter sampling trains used in stack testing programs. In fact, large

quantities of gases are captured, and the erroneously indicated “emissions” of condensable particulate matter can exceed the emission rates of filterable particulate matter.

The erroneous data provided by the existing EPA method suggest that some combustion sources contribute significantly to primary emissions of condensed PM<sub>2.5</sub> particulate matter. At the very least, this conclusion is premature because the existing emissions data for condensable particulate matter are extremely limited and are subject to significant errors that create higher-than-true values.

There is a need for an accurate air emission testing procedure to determine the true condensable particulate matter emissions from combustion sources and other types of stationary industrial sources. This report is limited to a summary of the problems with EPA Method 202.

## 2. Method 202 Description

In 1991, EPA promulgated Method 202 for the measurement of condensable particulate matter. The purpose of this method was to help regulatory agencies compile data concerning the primary emissions of condensable particulate matter and the possible impact of these primary emissions on PM<sub>10</sub> and PM<sub>2.5</sub> ambient air quality.

In developing Method 202, EPA decided to use water-filled impingers as a convenient and readily available means to cool the sample gas below vapor dewpoint levels and to collect the particulate matter. The impingers are located at the back end of conventional particulate matter sampling trains (e.g. Method 5, Method 201A, or Conditional Method 040<sup>1</sup>) used to measure filterable particulate matter. In fact, the only changes necessary to adapt the water filled impingers in filterable PM sampling trains for the additional duty of measuring condensable particulate matter is to use additional laboratory processing steps to recover and weigh the condensed particulate matter recovered from the water-filled impingers. Following the sampling period, the inorganic and organic materials are separated in an extraction step, and the quantities of each type of condensable material are weighed.

## 3. Method 202 Measurement Biases

Air emission testing experience since 1991 has demonstrated that it is not appropriate to use water-filled impingers to cool the gas stream for condensable particulate matter measurement at sources having SO<sub>2</sub>, NO<sub>2</sub><sup>2</sup>, or soluble organic compound<sup>3</sup> emissions. These gaseous contaminants can absorb in the impingers and chemically convert to form material counted as condensed particulate matter. These gas reaction products are termed “artifacts” and are not related to the release or formation of ambient particulate matter.

Scrubbing the kiln stack sample gas stream in ice-chilled water-filled impingers creates chemical conditions that are entirely non-representative of condensable particulate matter formation in

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<sup>1</sup> Conditional Method 040 is an extension of Method 201A to simultaneously measure both filterable PM<sub>10</sub> and filterable PM<sub>2.5</sub>.

<sup>2</sup> NO<sub>2</sub> is one of the two oxidized nitrogen oxides included as NO<sub>x</sub>. NO<sub>2</sub> is usually present at levels of 2% to 10% of the total NO<sub>x</sub> concentration in the sample gas stream passing through the Method 202 impingers. NO emitted from the stack converts to NO<sub>2</sub> over periods of hours; however, this atmospheric conversion process is not relevant to the Method 202 sampling train.

<sup>3</sup> Soluble organic compounds include alcohols, aldehydes, ketones, and organic acids.

dispersing plumes from stacks. Most of the Method 202 biases result in condensable particulate matter emission test results that are higher-than-true. One potential Method 202 issue that has been proposed could result in test results that are lower-than-true. A description of some of the potentially more significant problems affecting Method 202 accuracy and precision is provided below.

- Dissolution of sulfur dioxide and nitrogen oxides into water with subsequent oxidation to form sulfates and nitrates in the impingers
- Incomplete purging of dissolved sulfur dioxide prior to the analyses for condensed material in the impingers
- Penetration of submicrometer sized condensed particles through the impingers of the Method 202 sampling train
- Gas phase homogeneous reactions between ammonia and hydrogen chloride and/or between ammonia and sulfur dioxide in the cold, water-filled impingers
- Dissolution of soluble organic compounds into water

Dissolution and Chemical Reaction of Sulfur Dioxide and Nitrogen Dioxide Gases - Since Method 202 was promulgated, there has been considerable concern that absorption of soluble sulfur dioxide and nitrogen dioxide and subsequent reactions of these dissolved gases occur within the aqueous phase in the impingers (references 1, 2, 3, 4, and 8). These reactions are important because these gases are considerably more soluble in cold liquids than in warm liquids. The 32°F to 68°F temperatures of the liquid in the ice-cooled impingers provide an ideal environment for the collection of soluble inorganic gases.

Atmospheric reactivity studies summarized in the final edition of the Particulate Matter Air Quality Criteria Document (reference 5) indicate that there are a number of reaction mechanisms for converting dissolved sulfur dioxide (sulfite ion) to sulfuric acid. These studies indicate that the conversion rate of sulfur dioxide in water droplets can be “several times” higher than the 1% to 3% per hour conversion rate observed for dry, gas phase reactions.

“Chemical reactions of SO<sub>2</sub> and NO<sub>x</sub> within plumes are an important source of H<sup>+</sup>, SO<sub>4</sub><sup>-2</sup>, and NO<sub>3</sub><sup>-1</sup>. These conversions can occur by gas-phase and aqueous-phase mechanisms. ... For the conversion of SO<sub>2</sub> to H<sub>2</sub>SO<sub>4</sub>, the gas-phase rate in such plumes during summer midday conditions in the eastern United States typically varies between 1 and 3% per hour, but in the cleaner western United States rarely exceeds 1% hr<sup>-1</sup>.”

U.S. EPA, Particulate Matter Criteria Document (October 2004), Page 3-63.  
(Concerning dry, gas phase reactions)

“The contribution of aqueous-phase chemistry to particle formation in point-source plumes is highly variable, depending on the availability of the aqueous phase (wetted aerosols, clouds, fog, and light rain)... The in-cloud conversions of SO<sub>2</sub> to SO<sub>4</sub><sup>-2</sup> can be several times larger than the gas-phase rates given above.”

U.S. EPA, Particulate Matter Criteria Document (October 2004), Page 3-6.  
(Concerning aqueous phase reactions similar to those that could occur in Method 202 impingers)

Based on this general relationship, the aqueous phase conversion rates relevant to water filled impingers are estimated to be 2% to 6%. The secondary particulate (sulfate) formation rates at the aqueous phase conversion rates based on EPA research summarized in reference 5 are very high. Similar SO<sub>2</sub> oxidation rates in the impingers of Method 202 create significant quantities of sulfate material.

Incomplete Purging of Dissolved SO<sub>2</sub> - Method 202 is not silent regarding the SO<sub>2</sub> absorption and reaction issue. In fact, EPA recommends that the impinger solutions from the Method 202 sampling train be purged with clean nitrogen to strip out the dissolved SO<sub>2</sub> from the solution. Unfortunately, there are two factors that limit the effectiveness of this approach:

- (1) SO<sub>2</sub> reactions during the test run and prior to the start of the purge step and
- (2) incomplete purging of SO<sub>2</sub> from solution that ultimately oxidizes to form sulfates prior to the laboratory recovery of the Method 202 liquid samples.

Obviously, a post test run nitrogen purge has no impact on the quantity of SO<sub>2</sub> that reacts to form sulfuric acid in solution during the conclusion of the one-hour test run and the one to two hours after the test run that are often needed before purging is started. At a reaction rate of 2% to 6% per hour, there is considerable time prior to purging for the dissolved SO<sub>2</sub> to react to form sulfuric acid and subsequently be counted as “condensable particulate matter.”

It is also apparent that purging is more difficult than anticipated in Method 202. Tests conducted by Corio (reference 3) indicated that, in many cases, the one-hour purge time listed in Method 202 is not adequate to eliminate the gaseous material (e.g. sulfur dioxide) that has absorbed into solution. Similar problems have been observed by McCain and Williamson (references 2, 6, and 7). Purge efficiencies in the range of 80% to 90% have been measured in these tests. The remaining 10% to 20% of the dissolved sulfur dioxide (and other dissolved gases) is converted to condensable particulate matter in the samples in the hours to days that pass until the samples are analyzed. This can significantly affect the accuracy the test results.

To evaluate the extent of the error in Method 202 caused by SO<sub>2</sub> absorption and reaction, the quantity of SO<sub>2</sub> absorbed in the Method 202 impingers has been calculated based on Henry’s Law constants for a water - SO<sub>2</sub> solution (no dissolved carbon dioxide, hydrogen chloride, and/or sulfuric acid). The results are summarized in Table 1 for typical Method 202 sampling conditions. The calculations summarized in Table 1 are based on an SO<sub>2</sub> oxidation rate of 4% per hour, a gas sample volume of 30 DSCF, and a 3-hour period after the test run for the start of the nitrogen purge.

In reviewing these calculated biases, it is important to note that the sulfur dioxide oxidation reactions start at the very beginning of the test run when sulfur dioxide first enters the solution. Due to the minimum run time of one hour, the dissolved gas has time to oxidize to form a condensable species even if the testing company initiates the purge step immediately after sampling.

The hourly and annual artifact “emissions” are based on a total stack gas flow rate of 50,000 DSCFM and 7,500 operating hours per year. The stack gas flow rate is representative of a small-fossil fuel-fired boiler, a moderately sized kiln, or a moderately sized furnace.

It is important to note that the positive bias levels in Table 1 concern conditions when SO<sub>2</sub> is absorbed into a “clean” water solution. The research data cited by EPA in the Particulate Matter

Criteria Document (October 2004) indicate that the presence of ammonia significantly enhances the absorption of sulfur dioxide and its subsequent oxidation in the water droplets. This impact is due, at least in part, to the neutralization of acidic dissolved carbon dioxide by the very basic ammonium hydroxide. These conditions are directly relevant to the conditions present in Method 202 impingers sampling the effluent gas stream of a combustion source.

SO <sub>2</sub> , ppm	N <sub>2</sub> Purge Efficiency, %	Sulfuric Acid Artifact, grains/DSCF	Sulfuric Acid Artifact, Equivalent lbs/hour	Sulfuric Acid Artifact, Equivalent tons/year
200	No Purge	0.0157	6.7	25.2
	80	0.0055	2.3	8.8
	90	0.0039	1.7	6.2
	95	0.0031	1.3	5.0
	100	0.0023	1.0	3.7
500	No Purge	0.0449	19.3	72.2
	80	0.0136	5.8	21.9
	90	0.0097	4.2	15.6
	95	0.0078	3.3	12.5
	100	0.0058	2.5	9.3

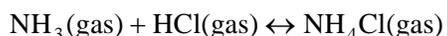
Laboratory tests conducted by Air Control Techniques, P.C. collaborate the positive bias due to SO<sub>2</sub> absorption and oxidation. In tests using clean impinger water, condensable particulate matter levels exceeding 0.014 grains/DSCF were found even though the Method 202 laboratory tests were conducted with a sample gas stream blended with Protocol 1 high quality particulate-free gaseous sulfur dioxide, nitrogen oxides, and ammonia (reference 4). There was no condensable particulate matter in the blended gas stream entering the Method 202 sampling train. The measured “condensable particulate matter” levels observed in these tests clearly demonstrate the significant positive biases that can exist with Method 202 techniques. The positive bias levels in Table 1 also do not include the error introduced by the retention of dissolved NO<sub>2</sub> and soluble organic compounds.

The positive biases in Table 1 do not reflect the impact of acid gases such as hydrogen chloride, sulfuric acid, or carbon dioxide in the sample gas stream. The presence of these gases would partially reduce sulfur dioxide solubility.

In addition to the positive biases discussed above, it is possible that Method 202 is biased to higher-than-true emission levels due to the conversion of sulfur dioxide and nitrogen oxides that occurs in the front half (Method 5, Method 201A, or Conditional Method 040) portion of the sampling train. This could occur due to contact of the gas stream with the hot metallic surfaces of the nozzle and the probe and/or contact with probe or filter deposits of particulate matter with catalytically active constituents on the particle surfaces. This catalytic reaction mechanism can potentially result in the formation of vapor phase sulfuric acid and nitric acid that would not form in the plume; however, there are insufficient data available to determine if this reaction mechanism is significant.

Penetration of Submicrometer-Sized Particles Through the Impingers - Some agency representatives have expressed concern that submicrometer sized particles formed in the Method 202 impingers are not collected at 100% efficiency in the impingers. There are no published studies presently available that confirm this negative bias or quantify the extent of this problem.

Gas Phase Homogeneous Reactions - The bias caused by certain gas phase reactions in Method 202 is well recognized and is acknowledged in Method 202. Gas phase reactions between ammonia and either hydrogen chloride or sulfur dioxide are very rapid in the less than 68°F environment of the Method 202 impingers. Atmospheric chemistry studies have shown that ammonium chloride forms in a concentrated gas stream when the air temperature is cold. The ammonium chloride then disassociates to reform gaseous ammonia and gaseous hydrogen chloride when the concentrations of each of the gases decrease during plume dispersion.



Gas Phase Reaction



Condensation/Evaporation

In Method 202, the ammonium chloride is trapped in the impingers, and the reverse reaction to form ammonia gas and hydrogen chloride gas is prevented. This results in the formation of measured condensable particulate matter that is not present as condensed particulate matter in the atmosphere.

Dissolution of Soluble Organic Compounds - Some combustion sources can have low-to-moderate concentrations of soluble organic compounds such as alcohols, aldehydes, ketones, and organic acids. These are efficiently absorbed in the cold aqueous solutions in the Method 202 impingers. The partial retention of these soluble organics can significantly bias the Method 202 test results.

#### **4. Summary of Method 202 Issues**

The positive biases relating primarily to the absorption and chemical reaction of gases inappropriately captured in a Method 202 sampling train can result in measured emission rates that are in the range of 0.005 to more than 0.015 grains/DSCF based on laboratory studies of Method 202 and solubility calculations similar to those discussed earlier. In some cases, the Method 202 biases can potentially be higher.

Calculated emission of condensable particulate matter based on the Method 202 artifact quantities incorrectly suggests emissions that are equal to or higher than the filterable particulate matter emission rates from combustion sources controlled with high efficiency particulate matter control systems. These very high positive biases might be partially offset by a negative bias caused by incomplete capture of submicrometer-sized particles in the Method 202 impingers.

Method 202 has been shown to be an inappropriate emission testing procedure for determining condensable particulate matter emissions from combustion sources. Emission source operators and regulatory agency personnel interested in accurate condensable particulate matter emissions data should postpone testing until a new and more reliable method has been developed and adequately verified. In the interim, if testing must be performed, it would be prudent to use a modified Method 202 sampling train or a prototype air dilution sampling train. The condensable

particulate matter emissions data obtained by use of Method 202 have significant errors that do not serve the interests of the public, the regulatory agencies, or the lime plant operators.

Alternative test methods should be developed to provide accurate condensable particulate matter data. Unfortunately, the one method being developed by EPA for this purpose, Conditional Method 039, continues to be subject to numerous practical problems. The method is extremely expensive, extremely complicated, and is basically not yet “field ready.” Alternative practical and economical methods will eventually be developed by ASTM or other standard-setting organizations.

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ATTACHMENT B:  
AP-42 Factors for Condensable PM from Lime Kilns

This analysis evaluates the basis of the condensable PM emission factors (lb/ton lime) for fabric filter-equipped rotary lime kilns, as follows<sup>1</sup>:

Source	Inorganic	EMISSION FACTOR RATING	Organic	EMISSION FACTOR RATING
Coal-fired rotary <u>preheater</u> kiln with multiclone, water spray, and fabric filter (SCC 3-05-016-22)	1.1	E	0.15	E
Coal-fired <u>straight</u> rotary kiln with fabric filter (SCC 3-05-016-18)	0.38	E	no data	

Source: AP-42, Table 11.17-2. Emission Factors for Lime Manufacturing Calcining, Cooling, and Hydrating, Feb. 1998.

All of the factors are E-rated. This rating is characterized by EPA as:

Poor. Factor is developed from C- and D-rated test data, and there may be reason to suspect that the facilities tested do not represent a random sample of the industry. There also may be evidence of variability within the source category population.

These emission factors are based on tests conducted between 1974 and 1977. All of the tests used the 1971 version of Method 5, with condensable PM collected using back-half impingers described as an option to determine the stack gas moisture content. Since that time, EPA has provided improved the procedures for condensable PM testing in Method 202. However, as explained in Attachment A, test results derived from using Method 202 likely have significant positive biases.

The **rotary preheater** EFs above reference a 1974 test at the Marblehead Lime plant in Indiana.<sup>2</sup> However, the tested kiln was a straight, not a preheater rotary kiln. Organic condensables are reported as “back-half organic extract,” a procedure that is not described in Method 5. Regarding the inorganic fraction, the EF of 1.1 lb/ton lime is highly suspect because it is nearly 3 times the EF for straight rotary kilns. Due to the greater inherent scrubbing of gases with lime in preheater kilns, such kilns would be expected to emit less (not more) condensable PM.

<sup>1</sup> Of the 15 BART-eligible lime kilns in VISTAS, 11 are fabric filter-equipped rotary kilns.

<sup>2</sup> *Air Pollution Emission Test, Marblehead Lime Company, Gary, Indiana*, Report No. 74 LIM 7, U. S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC, 1974. (AP-42 ref. 34) ([http://www.epa.gov/ttn/chief/old/ap42/ch11/s17/reference/ref\\_34c11s17\\_Feb1998.pdf](http://www.epa.gov/ttn/chief/old/ap42/ch11/s17/reference/ref_34c11s17_Feb1998.pdf))

The **straight rotary kiln** EF references six different tests. One of the tests was conducted at a stone crushing operation, not a lime kiln.<sup>3</sup> The median of the results from the other referenced tests<sup>4</sup> is one-third the reported EF of 0.38 lb/ton lime:

<i>Type of PM</i>	<i>AP-42</i>	<i>Referenced Tests</i>	<i>Ref. 20</i>	<i>Ref. 21</i>		<i>Ref. 22</i>	<i>Ref. 23</i>	<i>Ref. 33</i>
inorganic condensable PM (lb/ton lime)	<b>0.38</b>	<b>0.16 (median)</b>	0.14	0.55	0.26	0.17	0.11	0.12

As discussed above, the impinger methods used in these tests likely create significant overestimates of condensable PM.

<sup>3</sup> *Air Pollution Emission Test, J. M. Brenner Company, Lancaster, PA*, EPA Project No. 75-STN-7, U. S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC, November 1974 (AP-42 Reference 8)  
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<sup>4</sup> Reference 20: T. L. Peltier, *Air Pollution Emission Test, Martin-Marietta Corporation, Calera, AL*, (Draft), EMB Project No. 76-LIM-9, U. S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC, September 1975.  
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