Particulate Matter

The Impact of Winter NH_3 Emission Reductions on Inorganic Particulate Matter under Present and Future Regulated Conditions

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Extended Abstract

Atmospheric ammonia contributes to the formation of aerosol nitrate, an important constituent of inorganic fine particulate matter, and atmospheric ammonia is a significant source of acidification and excess nutrient loading to sensitive ecosystems. Recent regulation by the US Environmental Protection Agency requires large-scale emission reductions of NO_x and SO_2. These regulated emission changes will decrease the concentration of nitrate and sulfate and hence alter the composition of inorganic particulate mater.

The complexity of the inorganic aerosol system makes prediction of the importance of ammonia after these emission reductions non-trivial (Ansari and Pandis, 1998). Sulfate has a low vapor pressure and condenses to the aerosol phase. Any available ammonia gas will neutralize the sulfate to form ammonium sulfate aerosol. If additional ammonia is available in excess of what is needed to neutralize the sulfate, it can form aerosol ammonium nitrate in accordance with thermodynamic equilibrium.

The inorganic aerosol system has three states, listed in order of increasing ammonia concentration: (1) acidic, where there is insufficient ammonia to neutralize the sulfate, (2) ammonia limited, where all of the sulfate is neutralized, but the formation of ammonium nitrate is limited by scarce ammonia, and (3) nitrate limited, where ammonia is present in excess such that the formation of ammonium nitrate is limited by scarce nitric acid. Reductions in nitrate and sulfate will cause the system to become more nitrate limited, decreasing the sensitivity of inorganic $PM_{2.5}$ to NH_3. However, if the aerosol is initially acidic, then reductions in sulfate can cause some ammonia to become available for ammonium nitrate formation, which could increase the sensitivity of $PM_{2.5}$ to NH_3 emissions.

The change in ammonia deposition is tightly linked to the future state of the inorganic aerosol system. Aerosol ammonium has a longer lifetime (7-10 days) compared to gas-phase ammonium, which rapidly dry deposits (lifetime of one day) (Seinfeld and Pandis, 1998). Decreases in aerosol nitrate and sulfate may cause increases in gas phase ammonia and more ammonia will be deposited closer to emission sites.

To better understand the future role of ammonia as an atmospheric pollutant, we use the Community Multiscale Air Quality (CMAQ) Modeling System version 4.4 to simulate current and future regional air quality and deposition (Byun and Ching, 1999; Byun and Schere, 2005). CMAQ is an Eulerian grid model that simulates advection, dispersion, gas-phase chemistry, aerosol thermodynamics, cloud processes, and wet and dry deposition. The model domain consists of the continental United States, discretized into a 36km x 36km horizontal grid with 14 vertical layers. The meteorological inputs for January 2001 are derived from the Fifth Generation Penn State/NCAR Mesoscale Model (MM5) (Grell et al., 1994). We use the same meteorology to drive all of the emission scenarios and CMAQ simulations.

Ammonia emissions are from Gilliland et al. (*in press*). Other emitted species are from the National Emission Inventory 1999 version 3.0 grown to 2001 levels. The 2010 and 2020 year scenarios include regulated reductions in SO_2 and NO_x emissions as described in the Clean Air Interstate Rule and Clean Air Visibility Rule (USEPA, 2005). The emission changes from 2001 to the future scenarios for SO_2, NO_x, and NH_3 are listed in Table 1.

Table 1. Domain-average emission changes for January for each scenario.

Species	2010	2020
NH_3	+4%	+13%
SO_2	-28%	-35%
NO_x	-30%	-40%

To calculate the sensitivity of $PM_{2.5}$ to NH_3 emissions, we simulate both the base case emission scenario for 2001, 2010, and 2020, and we simulate each of these scenarios with a 10% reduction in NH_3 emissions. The emission reduction is calculated by multiplying by a fixed factor across all NH_3 emission sources, locations, and time periods. CMAQ is run for January 2001 for each of these six scenarios. The inorganic $PM_{2.5}$ (iPM) is the sum of NH_4^+, SO_4^{2-}, and NO_3^- concentrations. The sensitivity of inorganic $PM_{2.5}$ to NH_3 emissions is then calculated as

$$\Delta iPM \Big/ \Delta NH_3 \equiv \frac{(iPM_{1.0} - iPM_{0.9})}{(iPM_{1.0})}$$

where $iPM_{1.0}$ is the monthly-averaged inorganic $PM_{2.5}$ concentration for the base case emission scenario, and $iPM_{0.9}$ is the monthly-averaged inorganic $PM_{2.5}$ concentration for the emission scenario with a 10% decrease in ammonia emissions.

To evaluate the change in reduced-form nitrogen deposition due to changes in future scenario NO_x and SO_2 emissions, we examine the wet deposition, dry deposition, and the sum of wet and dry deposition from gas-phase NH_3 and aerosol NH_4^+ for the entire month at each grid cell. The difference is calculated between the 2001 and 2010/2020 scenarios.

When looking at select non-attainment sites for winter conditions in January, we find that the concentration of inorganic $PM_{2.5}$ is less sensitive to NH_3 emission reductions in the future scenarios. However, the reductions are relatively small and the inorganic $PM_{2.5}$ sensitivity to NH_3 emission reductions remains greater than zero. At locations harboring sensitive ecosystems, we find increases in total reduced-form nitrogen deposition near NH_3 emission sources. More of the total ammonia is in the gas phase which deposits more readily than aerosol ammonium.

Disclaimer

The research presented here was performed under the Memorandum of Understanding between the U.S. Environmental Protection Agency (EPA) and the U.S. Department of Commerce's National Oceanic and Atmospheric Administration (NOAA) and under agreement number DW13921548. This work constitutes a contribution to the NOAA Air Quality Program. Although it has been reviewed by EPA and NOAA and approved for publication, it does not necessarily reflect their policies or views.

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Concentrations of Nitrogen and Sulfur Species in the Gas and Particle Phases Downwind of Two Dairy Operations

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Abstract

Measurements of gas and particle phase nitrogen and sulfur species were made downwind of two dairy operations in eastern Colorado in 2005. The primary $PM_{2.5}$ ionic species, Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}, gas phase concentrations of HNO_3, NH_3, and SO_2, and species' size distributions were measured using cyclone/annular/filter pack systems and multi-stage impactors. A PILS (Particle Into Liquid Sampler)/IC (Ion Chromatograph) system was deployed for high time resolution (10-15 min) measurements of $PM_{2.5}$ composition to provide a better understanding of changes in aerosol concentrations and their relation to transport.

Preliminary analysis of data from the summer and winter 2005 campaigns reveals high concentrations of gas phase ammonia (from ~10-1000 $\mu g/m^3$) downwind of dairy operations. At both study locations more than 90% of the total N(-III) (particulate ammonium + gas phase ammonia) was present as gaseous ammonia. Approximately 40-60% vs. approximately 15% of the total N(V) (particulate nitrate + gas phase nitric acid) were present as nitric acid in summer and winter, respectively. The predominance of gas phase species was favored by hot, dry conditions. Additional observations of particulate species size distributions and temporal variability in species concentrations are also reported.

Introduction

Emissions of ammonia from confined animal feeding operations are a potential concern in terms of their impacts on fine particle concentrations of ammonium nitrate and sulfate. The combined presence of NH_3, H_2SO_4, and HNO_3 can form particulate ammonium sulfate ($(NH_4)_2SO_4$) and particulate ammonium nitrate (NH_3NO_3) through gas to particle conversion reactions (Richardson et al., 1987; Seinfeld et al., 1998). Ammonium nitrate formation is a reversible reaction, with an equilibrium constant dependent on temperature and relative humidity. Relatively high humidities and low temperature favor particulate ammonium nitrate formation. In the absence of sufficient ammonia to fully neutralize sulfate, formation of particulate ammonium nitrate is not favored (Lee et al., 2004). If excess ammonia is available, however, ammonium nitrate can form.

Methods

Commercially available URG cyclone/annular denuder/filter pack systems were used for $PM_{2.5}$ sampling (Figure 1). Ambient air is drawn through a cyclone (D_{50}=2.5 μm, 10 lpm), along two 242mm denuders in series, each coated with chemicals that absorb the gaseous species of interest. The remaining air stream is then filtered through a 2-stage filter pack using a Teflon filter for particle collection followed by a nylon filter to collect any volatilized nitrate. Particle samples were analyzed for Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+} and denuders were analyzed for gas phase concentrations of HNO_3, NH_3, and SO_2.

Figure 1. Typical URG sampler setup at upwind (left photo) and downwind (right photo) dairy locations.

Particle size distributions of key species were measured using a Micro-Orifice Uniform Deposit Impactor (MOUDI). The 8 main impactions stages of the MOUDI (Figure 2) collect the following aerodynamic diameter size ranges: (stage 1) 18 –10 µm, (stage 2) 10-5.6 µm, (stage 3) 5.6 – 3.2 µm, (stage 4) 3.2 – 1.8 µm, (stage 5) 1.8 – 1.0 µm, (stage 6) 1.0 – 0.56 µm, (stage 7) 0.56 – 0.32 µm and (stage 8) 0.32 – 0.18 µm and after-filter (<0.18µm). The MOUDI flow rate was 30 L/min. Total sample flow was measured by a pressure-corrected dry gas meter reading. Analysis of the collected size-resolved particulate matter will be focused on quantification of the primary ionic species (Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}).

Figure 2. MOUDI (left photo) and (PILS/IC) system (right photo)

Semi-continuous measurements of particle composition were made using a PILS (Orsini et al., 2003). The overall principle of PILS/IC is to collect particles that comprise the $PM_{2.5}$ aerosol mass into a small continuous flow of high purity water. This is done by condensing steam onto a denuded particle stream, followed by impaction of the droplets formed into a liquid stream. The liquid stream is drawn into two ion chromatographs for measurement of aerosol anions and cations. LiBr is used as an internal standard to account for steam dilution of the particle collection stream. High time resolution measurements using PILS/IC systems with switching between "gas + particle" and "particle only" sampling allows quantification of concentrations of major ionic species (Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) of

$PM_{2.5}$ and gaseous species (HNO_3, NH_3 and SO_2) with 10-20 minute time resolution (Figure 2). The "gas+ particle" system operates without denuders.

Results and Discussion

The average MOUDI size distributions measured at dairy operations in 2005 and 2006 are shown in Figure 3. Nitrate, ammonium, and sulfate were dominant ionic species in the fine particle mode. Nitrate is present as both fine mode ammonium nitrate and in the coarse mode, likely as calcium nitrate. Formation of particulate ammonium nitrate (NH_4NO_3) occurs by gas to particle conversion of NH_3 and HNO_3. Coarse mode calcium nitrate likely results from a reaction between gas phase nitric acid (or its precursors) and soil dust particles.

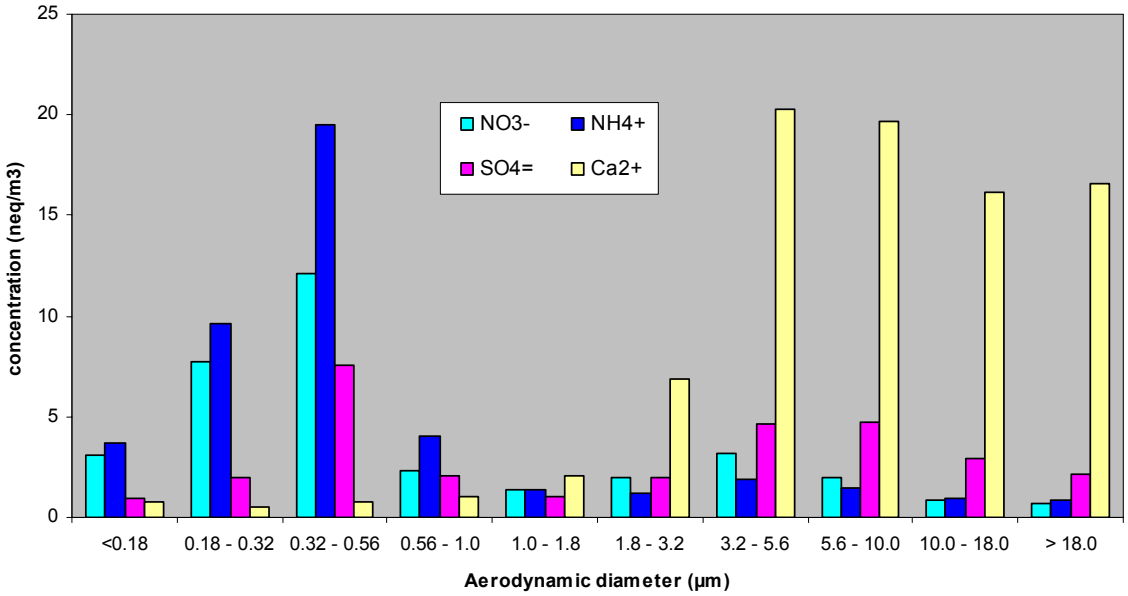

Figure 3. Average ion species' size distributions observed at two dairy operations.

Formation of particulate ammonium nitrate is not favored thermodynamically during warmer periods. While approximately 40-50% of total N(V) (particle nitrate + gas phase nitric acid) was present as nitric acid in July 2005, total N(V) tended to be associated mainly with particles during colder periods in November 2005 (Figure 4). More than 90% of the total N(-III) (particle ammonium + gas phase ammonia) was present as gaseous ammonia both in July and November 2005. The gas-particle phase distribution of sulfur species ranged between 70-90% gas phase in both warm and cool periods.

The PILS was deployed for several days at a dairy operation in November 2005 (Figure 5). Variability in major ion concentrations by wind direction is shown in Figure 6. High particle nitrate was observed during southerly flow, consistent with oxidation of NO_x emitted in urban areas to the south (Fort Collins and Denver are both south of the dairy) followed by particle formation resulting from reaction with gaseous ammonia.

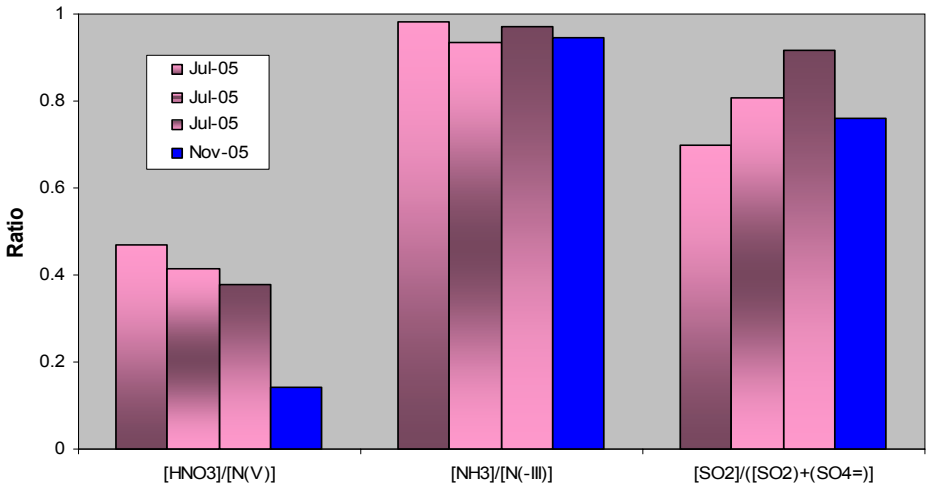

Figure 4. The ratio of HNO_3, NH_3 , and SO_2 to total N(V), N(-III), and ($[SO_2]+[SO_4^{2-}]$)

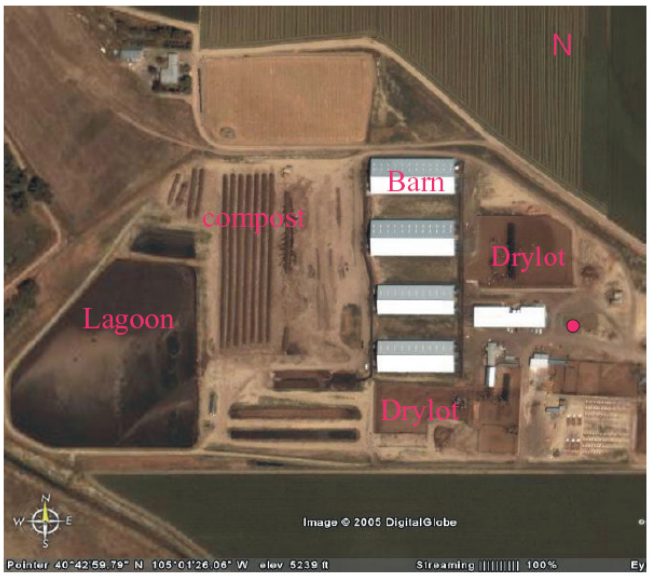

Figure 5. PILS measurement location (red dot) at a dairy studied in November 2005.

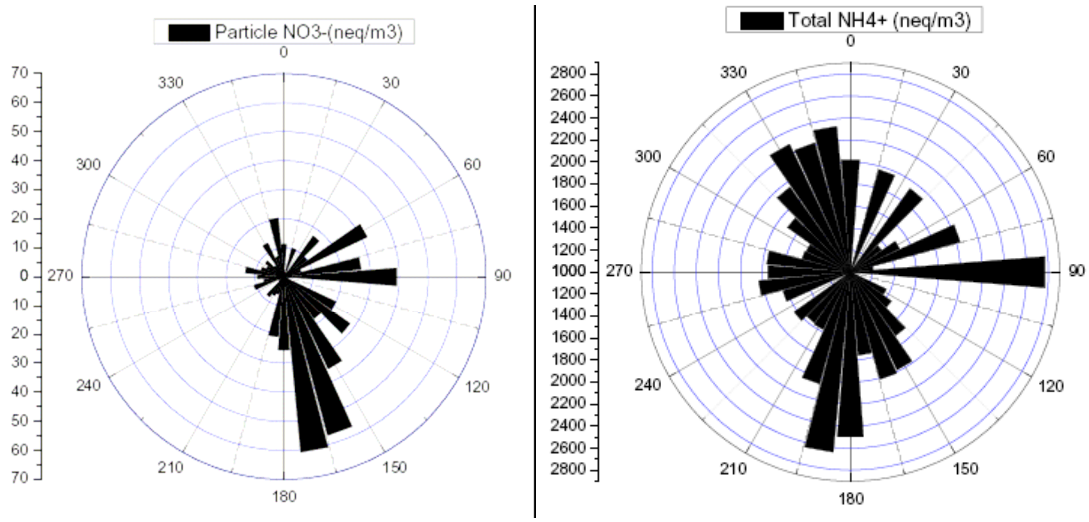

Figure 6. Particle nitrate and total ammonium from PILS with wind direction

PILS total N(-III) (gaseous ammonia + particulate ammonium) concentration variations with wind direction reveal varying contributions by different sources at the dairy. Higher concentrations were observed, for example, with flow from the northwest, consistent with emissions sources located in a large drylot and from barn areas. Comparison of total N(-III) measured by PILS with simultaneous N(-III) measurements with the URG sampler reveal considerable loss of gaseous ammonia within the undenuded PILS system, likely resulting from capture of ammonia by wet interior surfaces of the steam condensation chamber. Future efforts will make use of an alternative steam sampler design that is more efficient at total N(-III) collection.

Acknowledgements

Special thanks are given to Tim Hinerman and Alicia Evans for support and study planning. This work was funded by the USDA through the Colorado State University Agricultural Experiment Station.

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Agriculture and Air Quality – Airborne Particulate Matter

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Abstract

Air quality impacts from agricultural activities have generating considerable interest in the past decade. This increased interest stems from several reasons, including urban sprawl, which has led to closer proximity between urban populations and rural, farming areas; intensification of some agricultural activities, such as the increasing size of animal feeding operations; better understanding of secondary air pollutant formation, including secondary aerosols; and continuing revisions of national ambient air quality standards, including those for particulate matter (PM), reflecting better understanding of the effects of exposure to air pollutants. The U.S. Environmental Protection Agency has recently completed a review of the national ambient air quality standards (NAAQS) for PM and has proposed significant revisions to the primary and secondary NAAQS for PM. These changes, which include revision of the 24-hr PM2.5 standard, have resulted from new understanding of the role that fine particles (typically nominally smaller than 2.5 micrometers in aerodynamic diameter) play in health effects associated with short-term exposure. New standards are also proposed for a new indicator of thoracic particles – those particles with aerodynamic diameters between 2.5 and 10 micrometers (PM10-2.5) to target "urban" dust – i.e., resuspended road dust and industrial and construction sources, but excluding "rural" dust including windblown dust and PM from agricultural fields. This proposed PM10-PM2.5 standard would replace the current PM10 standard. A new, sub-daily fine particle standard is also under discussion. The agricultural sources of ambient particulate matter are reviewed, as well as their chemical and size characteristics, and what is known of their emission inventories.

Thoracic Particles

Thoracic particles (also commonly known as coarse fraction particles, or approximately PM10-2.5) are airborne particles that are generated from mechanical processes. Thoracic particles associated with agricultural activities are usually geologic in origin, and include windblown dust from fields, dust from rural roads, as well as dust generated by specific farming practices such as harvesting, plowing, or planting. Animal feedlots may also represent a source of thoracic particles. In these cases, the particles likely also contain bioaerosols, endotoxins, or allergens.

Windblown dust. Dust from dry fields especially during windy periods remains a concern for many agricultural areas in the US, particularly in arid regions. Windblown dust represents an aesthetic nuisance at best, and in worst cases has led to traffic fatalities due to significantly impaired visibility (Sundram et al., 2004). There is little current information to suggest that inhalation of windblown dust from agricultural fields represents a significant health risk, although reports of "dust pneumonia" were somewhat common during the Dust Bowl Era. There is some limited work that has suggested a link between exposure to coarse fraction particulate matter and respiratory ailments, although not specifically to agricultural coarse fraction PM. There have been several significant efforts to understand those conditions that lead to windblown dust, through monitoring and modeling activities. Some of these modeling efforts have included the Columbia Plateau PM10 Project in Central Washington, and the Wind Erosion Prediction System developed by scientists at the USDA. Modeling of windblown dust requires meteorological inputs, up-to-date land use information, terrain data, knowledge of surface treatment, and significant soil characterization including moisture, clay content, and amount of aggregation. Compounding the complexities of assembling this information is the need for detail at high temporal and spatial resolutions. Most modeling efforts have been conducted for very specific regions, and all are empirical or semi-empirical in nature. One significant exception is the WEPS, which takes into account detailed historical information such as the biomass both above and below ground, soil surface treatments, and precipitation history, and which tracks the resulting soil conditions in a chronological fashion.

Specific farming activities. Dust from a number of specific farming activities, such as almond harvesting or field plowing, has been studied, especially in California, in an effort to determine the contributions from these activities to particulate pollution there.

Animal feeding operations. Thoracic particles can be emitted from animal feeding operations (AFOs) through several mechanisms including animal activity, air circulation fans in housing units, entrainment of mineral and manure dust, and spraying of liquid wastes onto fields. These activities have not been extensively characterized for their PM emission potentials, although some studies, especially in Europe, are reported in the peer-reviewed literature, especially for dust potential in animal housing.

Measurement issues. Researchers particularly at TAMU have reported on oversampling problems that arise in the EPA Federal Reference Method (FRM) for particulate matter (Capareda et al., 2005). Oversampling occurs as a result of the confluence of particle size distributions and performance characteristics of the FRM PM10 sampler. PM10 samplers ideally would collect particles smaller than 10 micrometers in aerodynamic diameter with 100% efficiency, and particles larger than 10 micrometers would not be collected at all. In reality, however, some larger particles are also collected, and the PM10 performance is thus based upon collection of particles of 10 micrometers at 50% efficiency. Parnell and coworkers argue that agricultural PM is larger in particle size than typical urban PM10, so that the entire particle size distribution from agricultural activities is shifted toward larger particles. They have shown that this results in oversampling of PM10 when a FRM PM10 sampler is used. These researchers have developed a correction procedure that uses the ratio of PM10 to TSP to correct for the larger size distributions for various types of dust.

Emission inventories. Several compilations of emission factors have been produced, including the EPA document, AP-42, and the recent studies sponsored by the Western Governors Association – Western Regional Air Partnership (WGA-WRAP) Fugitive Dust studies.

Health questions. The EPA has recognized that exposure to thoracic particles generated from agricultural activities does not appear to represent a significant health risk, and so agricultural PM10-PM2.5 has been recommended for exemption from regulation. However, there remains other potential health issues that should be examined and which may be difficult to delineate from co-pollutants (other pollutant species that occur at the same time or from the same source). These include potential exposure to pathogens from blowing dust containing manure particles, endotoxins on bioaerosols from animal feeding operations (e.g., Rosas et al., 2001), as well as pesticides or metals carried on dust particles. In addition, there is some evidence to suggest that PM carries odor-causing species, thus making it difficult to separate odors from PM.

Fine Fraction PM

Primary fine particles from agricultural activities include smoke from field burning, as well as secondary aerosols formed in the atmosphere, for example from reactions involving gaseous ammonia or hydrogen sulfide from animal feeding operations.

Biomass combustion. Fire is used as a tool for managing field residues after harvest, weeds and pests, and wastes. Fire is also used in forestry management for the same reasons. Smoke from burning biomass may cause visibility problems and respiratory problems especially for certain vulnerable groups. As suburban areas sprawl toward farming areas, these problems become more commonplace. Smoke from biomass burning includes a number of air pollutants, including not only fine particles, but carbon monoxide, and a host of volatile and semivolatile organic compounds that include polycyclic aromatic hydrocarbons (PAHs) and methoxyphenols (MPs). Some PAHs have been found to be carcinogenic, while some MPs are respiratory irritants. While there has been some work to characterize both the chemical composition and emissions of PM and some PAHs and MPs in both vapor and condensed phases for some commonly burned biomasses (e.g., sugar cane residue, wheat residue, certain types of trees), this is an area that requires significantly more work. For example, recent studies conducted on Kentucky Blue Grass field stubble burning have shown significantly different PM emission factors and PAH and MP composition than similar studies conducted on wheat stubble burning.

Secondary agricultural sources. One of the least understood sources of agriculturally related PM is secondary aerosol – particles that result from condensation or chemical reactions between agriculturally

related gaseous species in the atmosphere. Probably the most important PM precursors from agriculture are ammonia and hydrogen sulfide (e.g., Baek et al., 2004). Significant sources of these two precursors are animal feeding operations of all kinds. Dairy farms, poultry farms, and pig farms in particular can be significant sources of precursors to PM.

Ammonia and hydrogen sulfide are emitted from wastewater lagoons, manure spraying operations, as well as manure piles (e.g., Aneja et al., 2000; Aneja et al., 2001). Emissions from lagoons have been relatively extensively studied, particularly as improvements in ammonia analyzing capabilities have been realized (e.g, Mount et al., 2002). Still, the high degree of variability between operations makes it difficult to quantify the extent of ammonia and hydrogen sulfide emissions from AFOs.

Measurement issues. Measurement issues for PM2.5 from agricultural activities are similar to those from other sources, and include organic artifacts from adsorption and/or desorption from filter media (e.g., Pang et al., 2002), and equilibrium issues especially with desorption of ammonium nitrate during sampling. In addition, researchers at TAMU also showed that there was a shift in the 50% cutpoint diameter for the PM2.5 FRM from the expected 2.5 micrometers to 2.7 $\pm$ 0.4 micrometers, so that the FRM form PM2.5 also oversamples agriculturally derived PM2.5 (Capareda et al., 2005).

Emission inventories. Emission factors for PM2.5 from fugitive dust including dust from paved and unpaved roads, construction sites, and windblown dust, as originally listed in AP-42, were developed using high-volume cyclone/impactor systems that have been shown to have a positive bias by as high as a factor of 2, compared to FRM-derived factors. Moreover, PM2.5/PM10 have sometimes been assumed to be constant but in fact for a variety of western soils this was not found to be true – rather, the ratio decreased with increasing PM10 concentrations. Emission factors from agricultural field burning have been quantified for a few crops, including wheat and Kentucky Blue Grass (Dhammapala et al., 2006, and references therein).

Health questions. Potentially hazardous compounds in agriculturally generated PM2.5 can include PAHs and methoxyphenols in biomass smoke (e.g., Roberts and Corkill, 1998). Because smoke from agricultural operations often impacts less populated areas, and because of the relatively short duration, but high concentration exposures, it has been difficult to demonstrate a statistically significant link between exposures and health effects, although recent epidemiological studies have been conducted in eastern Washington (Wu et al., submitted; Sullivan et al., submitted). Anecdotal evidence continues for an association between respiratory problems and smoke exposure, especially among people with asthma, and the practice remains controversial in various parts of the country (e.g., http://www.safeairforeveryone.com/).

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Modeling Approach to Estimate PM_{10} Emissions from Pig Husbandry

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Abstract

The level of measured dust emissions from livestock operations is generally determined by the number and weight of the animals as well as characteristics of the housing system, specified by management, animal species, feeding practices, bedding material, ventilation, as well as climatic inside and outside conditions. Modeling can be used to quantify the impact of management, seasonal, and diurnal variations. This approach combines measurements of the main influencing variables and estimated model parameters. The objective of the study was to estimate and model PM_{10} emissions from mechanically ventilated pig facilities.

Investigations were performed in mechanically ventilated pig houses for weaning, fattening, and sows in Italy and Germany. Thereby, the measurements included inside and outside concentrations of airborne PM_{10} particles (scatter light photometry), ventilation rate (calibrated measuring fans), and indoor air climate at a measuring frequency of 60 sec. Furthermore, feeding times and animal related data like weight and the (group) animal activity were recorded. Based on the sampled data, a multiple-input, single-output transfer function model was taken to describe the dynamic response of dust to variations of the measured variables.

As the results indicate, the average PM_{10} emission rate was influenced largely (> 100 %) by the specific housing system and/or management in the facility. Based on the three input variables ventilation rate, animal weight, and animal activity as well as model parameters, received on-farm, the minutely measured emission rate was simulated with a mean percentage error of 21 to 39 %. The simulated and measured average emission rate differed by about 4 % to 19 %.

Further recommendations of the study were to improve continuous and accurate measurements on the activity level in animal houses, to optimize the amount of measuring days in relation to the model accuracy, and to relate the modeling approach to intermittent measurements.

Introduction

Concentrations and emissions of dust particles from livestock operations are generally determined by the characteristics of the respective housing system, specified e.g. by management, feeding practices, bedding materials, number of animals, animal species, ventilation, as well as climatic inside and outside conditions (Takai et al., 1998).

Health impacts of aerosol particles depend mainly on their biological and chemical composition. Thereby, the origins of aerosols found in animal husbandry are mainly feed, bedding materials, skin, and excrements. Thus, they consist of a complex mixture made of organic dust as well as of relatively high concentrations of microorganisms, endotoxins, and NH_3 (Donham et al., 1986; Aarnink et al., 1999; 2004; Schneider et al., 2001; Seedorf, and J. Hartung; 2002; Takai et al., 2002). Such kinds of aerosols, which contain biologically active components and hence are likely to cause infections, allergic, toxic, or pharmacological reactions, are labeled in general as bio-aerosols (Cox, and Wathes, 1995). Aerosol particles emitting from animal

husbandry systems can be a nuisance towards the neighborhood, but can also be environmentally harmful (IPCC, 2005).

The emission rate of PM_{10} particles is in general determined by their release and dispersion from sources into the air, as well as by ventilation to the ambient. High ventilation rates during summer are accompanied in general with low indoor concentrations but increased emission rates, especially of inhalable and PM_{10} particle size fractions (Takai et al., 1998; Aerts et al., 2004; Jacobson et al., 2004; Koziel et al., 2004). Keck et al. (2004) investigated an increased impact of open yard exercises on the level of the absolute PM_{10} emission rate from pig husbandry during summer than during winter, as well as a significant influence on PM_{10} emissions with increasing growth stage of growing-finishing pigs. High peak-to-mean ratios during control activities of the farmers can be found especially in husbandry systems which use straw as bedding materials (Hartung, E. et al., 2004).

The influence of the animal activity on the dust concentration and emission is in general derived from observed differences between day and night-time and are related mainly to control activities of the farmers and feeding operations (Takai et al., 1998; Hinz and Linke, 1998; Wang et al., 2002), or it is derived from recorded characteristic daily patterns of the measured dust emission rate (Zeitler-Feicht et al., 1991; Koziel et al., 2004). Nevertheless, Gallmann (2003) and Schneider (2005) investigated also direct correlations between the animal activity and the PM_{10} concentration, both recorded at minutely frequencies. In general, it can be assumed that the dispersion of settled dust leads not only to a higher indoor dust concentration but also to an increase of the dust emission rate.

Modeling can be used to quantify the impact of management, seasonal and diurnal variations. This approach combines measurements of the main influencing variables and estimated model parameters. Hereby, the objective of the study was to estimate and model PM_{10} emissions from mechanically ventilated pig facilities.

Materials and Methods

Dust indoor concentrations and emissions were investigated in facilities for weaning, fattening and sows in Italy (Pianura Padana, Milan, Table 1) and Southern Germany (Hohenheim, Table 2). The investigated pig houses were equipped with mechanical ventilation systems each, separately controllable ventilation fans, and calibrated measuring fans. Temperature and humidity were measured and recorded via sensors of the ventilation control system and/or by additional sensors. Further recordings included the group animal activity in one pig facility, as well as the animal weight and feeding times in all investigated houses.

Measurements in Facilities for Weaning, Fattening and Sows in Italy

The measurements in Farm 1 in Italy (weaning and sows) included inside and outside concentrations of airborne PM_{10} particles (particles < 10 µm), ventilation rate, and indoor air climate with a measuring frequency of 1 min. Dust particles were sampled continuously ones per minute in the incoming air stream (weaning) as well as inside, close to one exhaust chimney (all facilities). Farm 2 (weaning) and Farm 1 (fattening) included measurements of PM_{10} concentrations at a frequency of 1 min; ventilation rate, outside and inside temperature, and humidity were recorded with a frequency of 15 min. In each of these facilities, the PM_{10} concentration was monitored optically using calibrated scatter light photometers (EPAM 5000, HAZ-Dust^{TM}; accuracy: +/- 3µg m^{-3}).

Temperature and humidity were recorded by the ventilation control system (FANCOM). Further, the measurements of the ventilation rate were performed with a high accuracy, separately for each exhaust chimney, using calibrated measuring fans (FANCOM; accuracy: +/- 45 m^3 h^{-1}; Berckmans et al., 1991).

The floor of the compartments was covered either with grooved plastic slats (weaning) or featured slatted concrete floor elements (fattening, sows). Feeding was applied dry or liquid, ad libitum or at three to four fixed feeding times per day (Table 1). A detailed description of the facilities is provided in Costa et al. (2004) and Costa et al. (2005).

Table 1. Investigated time periods and research facilities in Italy

	Number of animals	Average animal weight	Food supply and Feeding times	Floor material
Sows Farm 1, 18 Feb-25 Mar 04	24 to 43 sows	approx. 180 kg	dry (ad libitum)	slatted concrete floor
Weaning Farm 1, 25 Sep-9 Nov 04	350 piglets	7 kg to 36 kg	dry (ad libitum)	grooved plastic slats
Weaning Farm 2, 5 Sep-10 Oct 05	250 piglets	6-7 kg to 30 kg	liquid + dry (manually) 7:40; 13:40; 17:00	grooved plastic slats
Fattening Farm 1, 15-23 Nov 05	329 pigs	71 kg to 77 kg	liquid (automatically) 9:30; 12:30; 16:30; 19:30	slatted concrete floor

Measurements in the Fattening Pig Facility in Germany

Investigations in the research facility for fattening pigs in Germany were performed during two fattening periods (18 Nov 2003 until 24 Feb 2004 and 13 Apr 2004 until 20 Jul 2004) in two compartments. Thereof, dust concentrations were measured at four time periods (1080 min to 2550 min) per fattening period, either in compartment 1 or in compartment 2 (Table 2).

Airborne PM_{10} particles (particles < 10 µm) were monitored simultaneously at one representative measuring point indoors (all measuring periods), as well as in the exhaust shaft (six out of eight measuring periods). For the optical dust monitoring, calibrated scatter light photometers (TSI DustTrak 8520 ™; accuracy: +/- 1 µg m^{-3}) were used.

A vertical projection of the research facility and detailed table about the measuring instruments and accuracies for the measurements of temperature, relative humidity, ventilation rate, and animal activity is given in Haeussermann et al. (2006). Thereof, the group animal activity per pen (27 pigs) was recorded with infrared sensors (Pedersen and Pedersen, 1995), the ventilation rate was measured using calibrated measuring fans (MULTIFAN; accuracy: +/- 20 m^3 h^{-1}; Hartung, E., 2001). The measuring frequency for dust, ventilation rate, indoor temperature, humidity, and animal activity was one average value per minute.

The two pens per compartment featured a concrete slatted floor. Straw was supplied weekly via an occupation equipment, type "Porky Play" (straw capacity: approx. 3-4 kg). Feed was provided either liquid or in mash form (Table 2). Fresh air was coming in via two air inlet pore channels per compartment, the outgoing air was extracted under-floor (Haeussermann et al., 2006).

Table 2. Investigated time periods in the research pig facilities in Germany

	Number of animals	Average animal weight	Food supply and Feeding times	PM_{10} measurements
12-13 Dec 2003 09-11 Feb 2004 25-26 May 2004* 29-30 Jun 2004	53 pigs 53 pigs 53 pigs 53 pigs	50 kg 101 kg 61 kg 90 kg	Liquid (sensor) automatically 20 feeding times: 6:00 until 22:00)	Indoors[1] & exhaust shaft
22-23 Dec 2003 21-22 Jan 2004 26-27 Apr 2004 08-09 Jun 2004	54 pigs 54 pigs 54 pigs 54 pigs	58 kg 85 kg 35 kg 73 kg	mash (ad libitum) automatic food supply at six feeding times: 6:10 until 20:10	Indoors[1] - Indoors[1] & exhaust shaft

* no measurements of temperature, humidity, ventilation rate, and animal activity (25-26 May 2004)
[1] control corridor, height: 1.70 m

Modeling Approach and Model Validation

The aim of modeling in this study was to simulate PM_{10} emissions, based on measurements of the main influencing variables and estimated model parameters. The dynamic response of the dust concentration and emission to variations of the measured variables was described by a multiple-input, single-output transfer function model (Young, 1984). The general structure of the used model is exemplified for a single-input by the following equation (Aerts and Berckmans, 2004):

$$y(k) = \frac{B(z^{-1})}{A(z^{-1})} u(k) + \xi(k) \quad (1)$$

where *y(k)* is the output, either of the dust concentration, mg m^{-3} or the dust emission rate, g h^{-1} at time *k*; *u(k)* is the respective input of the measured variable at time *k.* Thereby, input variables were selected based on their correlation with the PM_{10} concentration (Table 4 and Table 5) and via a multiple-input regression analysis. $\xi(k)$ is additive noise, assumed to be a zero mean, serially uncorrelated sequence of random variables with variance σ^2, accounting for measurement noise, modelling errors and effects of unmeasured inputs to the process. $A(z^{-1})$ *and* $B(z^{-1})$ are two series given by:

$$A(z^{-1}) = 1 + a_1 z^{-1} + a_2 z^{-1} + ... + a_{na} z^{-na} \quad (2)$$

$$B(z^{-1}) = b_0 + b_1 z^{-1} + b_2 z^{-2} + ... + b_{nb} z^{-nb} \quad (3)$$

where: a_j, b_j are the model parameters to be estimated; z^{-1} is the backward shift operator, with $z^{-1}.y(k) = y(k-1)$; *y* and *k* are defined as in Equation (1); *na*, *nb* are the orders of the respective polynomials. The model parameters were estimated in discrete time using the simplified recursive instrumental variable approach (Young, 1984). Time step of the model was one value per 60 sec.

The modeling approach was tested on four data sets of the research facility for fattening pigs, Germany (Table 2). Selection criteria for the data sets were complete data, including indoor and exhaust PM_{10} concentrations, ventilation rate, temperature, humidity, and animal activity. The data set of 26-27 Apr was not selected due to considerable differences in the relations between dust and influencing variables, compared to the rest of the data sets. These differences were mainly explained by the early measuring period (fattening day 13-14) and related disturbances. Thus, a limitation of the model was given for the period between fattening day 24 (50 kg average weight; 12-13 Dec) and fattening day 85 (101 kg average weight; 09-11 Feb).

The model parameter estimation was performed on three of four data sets at each time, the validation of each model took place at the respective data set, which was not taken to build the model (Table 6). Validation criteria were (i) the ability to simulate the average measured dust concentration and emission of the validation set, and (ii) the agreement of the minutely measured and simulated values, expressed by the coefficient of determination R^2 and the root mean squared error (RMSE).

Results and Discussion

Variation of the Dust Concentration and Emission

The average PM_{10} indoor concentration of the different pig facilities varied from 0.11 to 0.73 mg m^{-3} (Table 3). Thereby, the lowest PM_{10} concentration occurred in the research facility for weaning pigs in Italy; the highest concentration was found in the research facility for fattening pigs in Germany. Compared to these minimum and maximum average values, the mean PM_{10} concentration at the three remaining facilities (weaning Farm 2, fattening pigs and sows, Farm 1, Italy) featured a narrower range of 0.33 to 0.48 mg m^{-3} (Table 3).

According to the dust indoor concentration, also the average PM_{10} emission rate featured a high variation throughout the stables for sows, weaning, and fattening pigs, which were in a range between 0.7 and 6.0 g d^{-1} LU^{-1} (Table 3; 1 LU = 500 kg). Similar to these investigations, the mean PM_{10} emission rates reported in literature, considering facilities for fattening pigs, varied from 0.8 to 4.3 g d^{-1} LU^{-1} (Götz, 2003; Aerts et al., 2004; Jacobson et al, 2004; Koziel et al., 2004). PM_{10} emission rates investigated by Jacobson et al. (2004) in gestation and dry sow barns were slightly higher and ranged from 0.5 to 9.1 g d^{-1} LU^{-1}.

In principle, feeding operations, the level of the animal activity, the ventilation rate, the indoor temperature, the animal weight and/or the fattening day, the housing system, as well as the management in the facility exerted an influence on the level of the dust indoor concentration and dust emission rate (Table 4; Table 5).

Thereby, the difference in the average PM_{10} emission rate between the research facilities for weaning and fattening in Italy and Germany (Table 3) can be associated on the one side with the higher ventilation rate per LU at the latter, and hence mainly due to seasonal difference. On the other side, the lower average

weight per animal in the weaning stable, in combination with differences in the farm management, led to a very low dust production, which was mirrored also by a relatively low indoor concentration at this farm (Table 3). Similarly, Gallmann (2003) reported a mean PM_{10} indoor concentration of 0.17 mg m^{-3} in a free ventilated kennel housing system for fattening pigs, during measurements outside the kennels, where the air quality is only minor influenced from the animals. Hereby, the ambient dust concentration outside the weaning farm in Italy averaged on 0.02 mg m^{-3} with a maximum value of 0.17 mg m^{-3}.

Table 3. Ventilation rate, PM_{10} indoor concentration and PM_{10} emission rate at the research facilities for sows, weaning and fattening pigs, Italy and Germany

	Ventilation rate [m^3 h^{-1} LU^{-1}]	Indoor concentration [mg m^{-3}]	Emission [g d^{-1} LU^{-1}]
Sows Farm 1, Milan 18 Feb-25 Mar 04	**96**[1] (63-308)[2] (20-577)[4]	**0.33**[1] (0.15-0.81)[2] (0.00-10.6)[4]	**0.70**[1] (0.26-1.75)[2] (0.00-18.0)[4]
Weaning Farm 1, Milan 25 Sep-9 Nov 04	**273**[1] (175-387)[2] (132-650)[4]	**0.11**[1] (0.06-0.17)[2] (0.00-3.23)[4]	**0.71**[1] (0.40-1.09)[2] (0.01-23.2)[4]
Weaning Farm 2, Milan 5 Sep-10 Oct 05	**245**[1] (160-328)[2] (81-827)[4]	**0.40**[1] (0.25-0.50)[2] (0.10-16.5)[4]	**2.38**[1] (1.20-3.60)[2] (0.43-24.2)[4]
Fattening Farm 1, Milan 15-23 Nov 05	**174**[1] (141-230)[2] (105-324)[4]	**0.48**[1] (0.32-0.65)[2] (0.02-5.61)[4]	**2.04**[1] (1.35-2.71)[2] (0.52-17.0)[4]
Fattening Farm, Hohenheim 12 Dec-30 June 04*	**481**[1] (235-854)[3] (189-1070)[4]	**0.73**[1] (0.35-1.26)[3] (0.06-6.41)[4]	**5.99**[1] (3.29-9.54)[3] (0.58-29.8)[4]

[1]mean [2]range diurnal means [3]range mean values data sets (*cf. Table 2) [4] total range
LU: Livestock Unit [1 LU = 500 kg animal weight]

Influences on the Dust Concentration and Emission

Correlation coefficients for the main influencing variables on the dust indoor and exhaust concentration in the research facilities for fattening pigs are demonstrated in Tables 4 and 5. In general, only low to negligible correlations occurred between the highly dynamic varying values of the minutely measured dust concentration and influencing variables like temperature, ventilation rate, animal activity, total weight, feeding times, and indoor humidity (Table 4 and Table 5). Nevertheless, the exhaust dust concentration at the research facility for fattening pigs, Germany, was explained with a regression coefficient of $R^2 = 0.61$ when taking into account all influencing variables listed in Table 4, except for the PM_{10} indoor concentration. In order to reduce the number of input variables, a sensitivity analysis was performed, taking into account either:

- indoor humidity, indoor temperature and/or ventilation rate,
- animal activity or feeding time,
- total animal weight or fattening day.

In conclusion, including the ventilation rate, the indoor humidity, the animal activity, and either the fattening day or the total weight of the animals as input variables resulted already in an R^2 of 0.61 and 0.60. Without indoor humidity, the regression coefficient was slightly lowered on $R^2 = 0.59$. In comparison, keeping the indoor temperature in the model but eliding the ventilation rate or the indoor humidity reduced the regression coefficient clearly to $R^2 = 0.56$. When replacing the animal activity by feeding times, the regression coefficient was reduced furthermore on $R^2 = 0.43$ if all other input variables were included and to $R^2 = 0.37$ if only the ventilation rate, the indoor humidity, and the weight of the animals were used in addition. Thus, in a first step, the animal activity, the ventilation rate, and the animal weight were taken into account for building a transfer function on the dust concentration and emission (Table 6).

Table 4. Correlation coefficients, research facility for fattening pigs, Hohenheim

	PM_{10} exhaust concentration	Dust indoor concentration	Tempe-rature	Relative humidity	Ventilation rate	Animal activity	total weight
PM_{10} indoor con.	0.943 **						
Temperature	- 0.435 **	- 0.420 **					
relative humidity	0.075 **	0.130 **	- 0.292 **				
Ventilation rate	- 0.314 **	- 0.332 **	0.682 **	- 0.501 **			
Animal activity	0.483 **	0.543 **	0.151 **	0.185 **	- 0.007		
Feeding time	0.145 **	0.159 **	0.008	0.027 *	0.010	0.220 **	
total weight	0.357 **	0.298 **	- 0.338 **	- 0.627 **	0.260 **	- 0.104 **	
Fattening day	0.310 **	0.250 **	- 0.270 **	- 0.659 **	0.344 **	- 0.119 **	0.995 **

significance: * $p < 0.05$ ** $p < 0.01$

A similar tendency for the correlation coefficients of the main influencing variables and the dust concentration like for the fattening pig facility in Germany, described above, was investigated at the research facility for fattening pigs in Italy (Table 5). Nevertheless, the indoor temperature and ventilation rate featured here a positive correlation with the dust concentration, the indoor humidity was correlated negatively instead. As the animal activity was not monitored, the maximum correlation resulted here in a regression coefficient of $R^2 = 0.23$, either by including all five available input variables or by taking into account the ventilation rate, the indoor humidity, the feeding times and the fattening day only.

Thereby, the influence of the ventilation rate on the dust concentration has to be regarded in general differentiated. During short time periods, an increase of the air exchange in the room can disperse settled dust and thereby result in an increased concentration of airborne particles. In addition, the effect of an increased ventilation rate on the spatial dust concentration and dust distribution depends largely on the respective airflow pattern in the building (Wang et al., 2002). A positive correlation between the ventilation rate and the dust concentration was found both in the fattening pig facility in Italy as well as in three of the four data sets of the research facility for fattening pigs in Germany when considering them separately. However, if the ventilation rate is clearly increased for a longer period, settled dust will be transported to the outside and a negative correlation is found between the ventilation rate and the concentration of airborne dust (Table 4).

Table 5. Correlation coefficients, research facility for fattening pigs, Farm 1, Milan

	Dust indoor concentration	Temperature [°C]	Relative humidity [%]	Ventilation rate [$m^3\ h^{-1}$]	Total weight [LU]
Temperature	0.261 **				
relative humidity	- 0.231 **	0.184 **			
Ventilation rate	0.272 **	0.947 **	0.212 **		
Feeding times	0.237 **	0.131 **	0.046	0.127 **	
total weight / Fattening day	0.129 **	- 0.533 **	- 0.590 **	- 0.525 **	1

significance: ** $p < 0.01$

First Results on Modeled Dust Concentration and Emission

As a first result of the model validation, the dynamic modeling approach enabled to simulate the mean dust indoor and exhaust concentration in tendency (Table 6). The average emission rate was simulated with an error of 4 % to 19 %, compared to the measured mean values. Thereby, the mean percentage error of the minutely simulated dust emission was in a range of 21 % (Figure 1) to 39 % (Figure 2). Although the dynamic course of the dust concentration and emission was simulated rather good on the data set of 12-13 Dec 2003 (Figure 2), which resulted in an R^2 of 0.70 to 0.77, the difference between the measured and the simulated mean was highest for this data set. Thereby, the difference between the measured and simulated mean at this validation was mainly due to the missing information about the influence of the low animal weight when estimating the model parameters from the three other data sets (Table 6). In contrary, the simulation of the mean dust concentration and emission was clearly improved for the two validation sets in June 2004 (Figure 1: 8-9 June). For both data sets the transfer function, built upon the respective three other data sets, included the information of the total variation of the ventilation rate and the animal weight, thus enabled a rather good simulation of the mean level of the respective validation set.

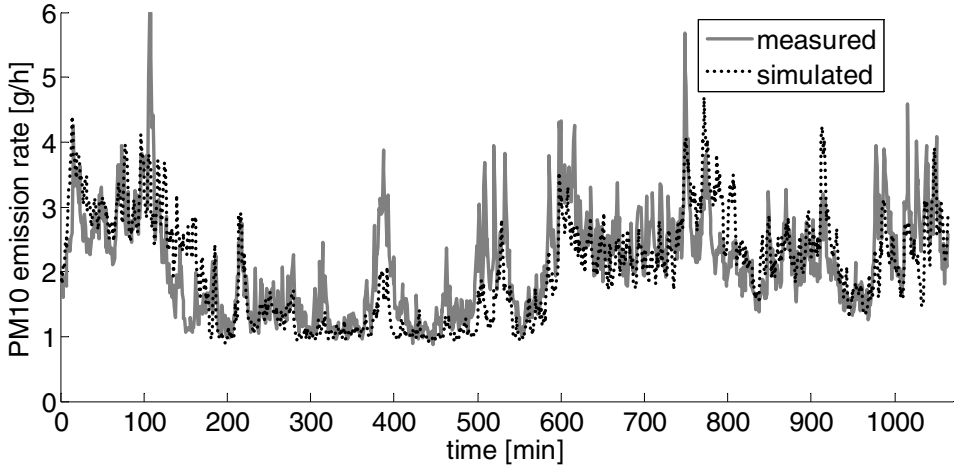

Figure 1. Measured and simulated PM_{10} emission rate, 8-9 June 2004 (54 pigs, average weight: 73 kg, mash feeding, ad libitum); input variables: ventilation rate, animal activity, and animal weight (Mean measured/simulated: 2.13/2.05, RMSE: 0.60, R_2: 0.529)

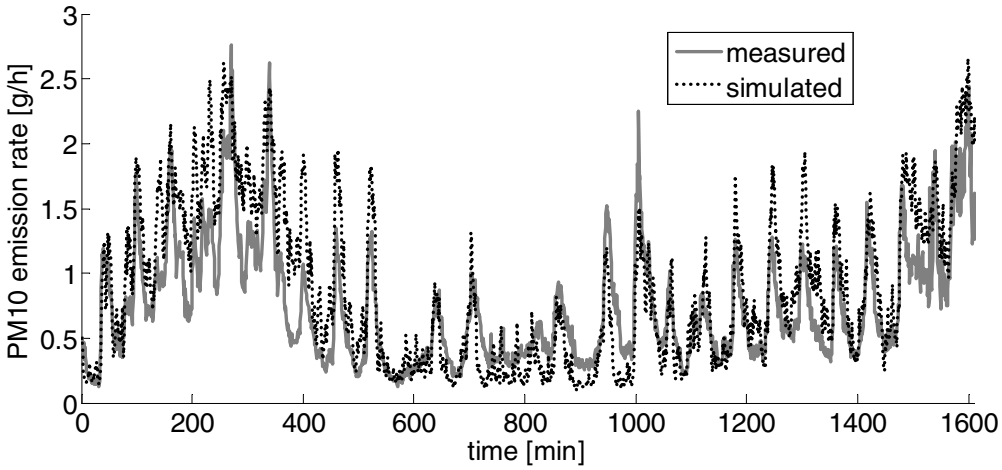

Figure 2. Measured and simulated PM_{10} emission rate, 12-13 Dec 2003 (53 pigs, average weight: 50 kg, sensor-liquid feeding); input variables: ventilation rate, animal activity and animal weight (Mean measured/simulated: 0.75/0.89, RMSE: 0.34, R_2: 0.772)

Table 6. Validation* (leave-one-out) of the transfer function model on PM_{10} concentration and emission, research facility for fattening pigs, Hohenheim (Input variables: ventilation rate, animal activity and animal weight)

	12-13 Dec 2003 (liquid feeding)	9-11 Feb 2004 (liquid feeding)	8-9 Jun 2004 (mash feeding)	29-30 Jun 2004 (liquid feeding)
Animal weight [LU]	5.3	10.7	7.9	9.7
ventilation rate [m^3 h^{-1} LU^{-1}]	mean: 371 *range: 246 – 649*	mean: 243 *range: 189 – 429*	mean: 854 *range: 796 – 907*	mean: 752 *range: 550 – 833*
PM_{10} Indoor concentration [mg m^{-3}]	**0.57[1] / 0.72[2]** *+- 0.29[3]*	**1.23[1] / 1.01[2]** *+- 0.65[3]*	**0.35[1] / 0.39[2]** *+- 0.29[3]*	**0.38[1] / 0.39[2]** *+- 0.20[3]*
	R^2: 0.702	R^2: 0.517	R^2: 0.351	R^2: 0.613
PM_{10} exhaust concentration [mg m^{-3}]	**0.37[1] / 0.52[2]** *+- 0.19[3]*	**0.79[1] / 0.58[2]** *+- 0.41[3]*	**0.31[1] / 0.30[2]** *+- 0.16[3]*	**0.30[1] / 0.33[2]** *+- 0.15[3]*
	R^2: 0.720	R^2: 0.482	R^2: 0.544	R^2: 0.442
PM_{10} emission rate [g h^{-1}]	**0.75[1] / 0.89[2]** *+- 0.34[3]*	**2.37[1] / 2.11[2]** *+- 1.07[3]*	**2.13[1] / 2.05[2]** *+- 0.60[3]*	**2.22[1] / 2.34[2]** *+- 0.84[3]*
	R^2: 0.772	R^2: 0.575	R^2: 0.529	R^2: 0.483

* Model is build out of three data sets, validation is performed at the respective fourth data set
[1] measured / [2] simulated; [3]RMSE: root mean squared error; LU: Livestock Unit [1 LU = 500 kg]

In order to improve the simulation of the PM_{10} concentration and emission rate, main future requests are:

- to optimize the database that is taken to build the model, including the total variation of the respective input variables,
- to perform measurements on input variables like the animal activity at a high time frequency and at a high spatial accuracy.

Conclusions

The average PM_{10} emission rate, measured in stables for sows, weaning and fattening pigs in Italy and Germany, featured a high variation and ranged between 0.7 and 6.0 g d^{-1} LU^{-1}, but was in general in accordance with PM_{10} emission rates reported in literature.

Main influencing variables on the PM_{10} concentration and emission were the ventilation rate, the (group) animal activity, feeding operations, the indoor humidity, the weight of the animals and/or the fattening day, as well as the housing system and the management in the facility. Taking these variables into account, the minutely measured PM_{10} exhaust concentration was explained with a regression coefficient R^2 of 0.60.

Hereby, the ventilation rate featured a negative correlation with the dust concentration in one building, while in another building a positive correlation occurred. We hypothesized that this is due to different airflow patterns in the buildings, as well as due to the length of the respectively considered time period.

The PM_{10} emission rate was modeled using a multiple-input, single-output transfer function model. Based on three input variables – ventilation rate, animal weight, and animal activity – the simulated and measured average emission rate of the respective validation sets differed by about 4 % to 19 %. Thereby, the mean percentage error of the minutely measured and simulated dust emission was in a range of 21 to 39 %.

However, further improvements of the modeling approach are necessary in order to realize an accurate simulation of the PM_{10} emission rate from pig facilities. This includes measurements on the activity level in animal houses at a high time frequency and at a high spatial accuracy.

Acknowledgements

The project was partly funded by the EU in the framework of a Marie Curie training site (reference number: HPMT-GH-01-00383-06).

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Particulate Matter Sampler Errors due to the Interaction of Particle Size and Sampler Performance Characteristics: Method 201a Stack Samplers

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Abstract

Agricultural operations are encountering difficulties complying with current air pollution regulations for particulate matter (PM). These regulations are based on the National Ambient Air Quality Standards (NAAQS), which set maximum concentration limits for ambient air PM. Source sampling for compliance purposes require the use of EPA approved samplers. Ideally, these samplers would produce accurate measures of the pollutant; for instance, PM_{10} samplers would produce accurate measures of PM less than or equal to 10 m (true PM_{10}). However, samplers are not perfect and errors are introduced due to established tolerances for sampler performance characteristics and the interaction of particle size and sampler performance characteristics. This paper focus on the inherent errors associated with the EPA approved PM_{10} stack sampler, sampler used in EPA's Method 201a sampling protocol. Results from this study show that if this sampler is used to sample PM with a mass median diameter (MMD) less than 10 microns then sampler would produce a concentration equal to the true concentration or if the MMD of PM being sampled was less than the cutpoint of the sampler then the sampler would begin to underestimate the true PM_{10} concentration. However, if the PM_{10} stack sampler were used in rural setting and was used to sample dust with a MMD of 20 microns and a geometric standard deviation of 1.5, then the sampler would overestimate the true PM_{10} concentration by 4.8 times. This sampler error is a consequence of the larger size PM associated with rural sources. It results in a substantial difference in the regulation of PM between urban and rural sources. To achieve equal regulation among differing industries, PM_{10} measurements MUST be based on true measurements.

Using Global Model and Satellite Data for Air Quality Studies

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Abstract

This study presents our global model analysis to estimate contributions of pollution, forest and agriculture fires, and long-range transport of aerosols to the surface PM2.5 concentrations in the U.S. and to interpret relationships between satellite observations of column aerosol optical thickness (AOT) and surface measurements of PM2.5 concentrations. Our model study for 2001 indicates that in the eastern half of the U.S., regional anthropogenic emissions, natural aerosols, and long-range transport of anthropogenic aerosols from other regions contribute about 62%, 32%, and 6% to surface PM2.5, respectively, while in the western U.S. the corresponding percentages are 28%, 60%, and 12%. Over 70 to 90% of sulfate aerosols over the U.S. are from the regional/local anthropogenic sources. Analysis of the relationship between the satellite AOT data from MODIS and surface PM2.5 concentrations from the IMPROVE network measurements reveals that the AOT and PM2.5 are much more closely correlated in the eastern than in the western U.S., mainly because of more uniform composition and less variable vertical profiles over the eastern U.S., as the model suggested.

Introduction

PM2.5 (particulate matter, or aerosol particles, with diameter less than 2.5 μm) is a key component determining air quality. At high concentrations, PM2.5 is harmful to human health, reduces visibility, and affects crop yields. Sources of PM2.5 include regional fossil fuel/biofuel combustions from industrial processes and transportation, fire emissions from agriculture activities and forests, and long-range transport from other regions. There are significant socio-economic benefits of accurate PM2.5 air quality forecasts, e.g., issuing health alert, providing guidelines for air quality management planning. Accurate PM2.5 forecasts require continuous extensive spatial and temporal monitoring of the current sates.

Remote sensing capability of atmospheric aerosol optical thickness (AOT) could lead to a quantum leap in our ability of air quality monitoring and prediction, especially for regions where surface monitoring network does not exist. In a recent EPA-NASA-NOAA prototype study, it has shown that the satellite AOT data from the MODIS instrument on the NASA EOS-Terra satellite closely track the surface PM2.5 concentrations over the U.S., providing an opportunity of "monitoring air quality from space" (Al-Saadi et al., 2005). On the other hand, studies have also shown that the correlations between AOT and PM2.5 vary with locations; they are much highly correlated in the eastern U.S. than in the western (Engel-Cox et al., 2004; Al-Saadi et al., 2005), revealing a challenge in quantitative use of satellite data for routine air quality prediction.

In this study, we use the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model to investigate the origin of surface PM2.5 and its relationship to total column AOT. We address the following questions:

- What are the contributions of anthropogenic emission, biomass burning, and long-range transport to the surface PM2.5 over the U.S.?
- How are the AOT and PM2.5 correlated, and how does this relationship change with location and time?
- How can we use the satellite data for PM2.5 studies?

The GOCART Model

The GOCART model is a global scale chemistry and transport model driven by the assimilated meteorological fields from the Goddard Earth Observing System Data Assimilation System (GEOS DAS).

Spatial resolution of the model in this study is at 2° latitude by 2.5° longitude and 30 vertical layers. GOCART simulates major aerosol types of sulfate, dust, black carbon (BC), organic carbon (OC), and sea-salt, and precursor gas species (e.g. SO_2 and dimethyl sulfide). Emissions from anthropogenic, biomass burning, biogenic, and volcanic sources and wind-blown dust and sea-salt are included in the model. Other processes include chemistry, convection, advection, boundary layer mixing, dry and wet deposition, and gravitational settling. Aerosol particle sizes from 0.01 to 10 µm are simulated with parameterized hygroscopic growth which is a function of relative humidity. Details of the GOCART model are described in our publications (Chin et al. 2000a, 2000b, 2002, 2003, 2004; Ginoux et al., 2001, 2004).

Comparisons of PM2.5 from GOCART with Observations

Figure 1 shows GOCART calculated daily aerosol species concentrations at the surface that are compared with measurements at 3 monitoring sites from the Interagency Monitoring Protected Visual Environments (IMPROVE) network for year 2001. Here, PM2.5 from the model is defined as the collection of sulfate, BC, OC, and dust and sea-salt particles with diameter less than 2.5 µm. Influence of long-range transport of sulfate and dust in spring is evident from both model and observations, and the peaks in August in the western high latitude site (Gates of the Mountains) are from biomass burning. Overall comparison of monthly averaged PM2.5 values for 2001 at 168 IMPROVE sites are shown in Figure 2.

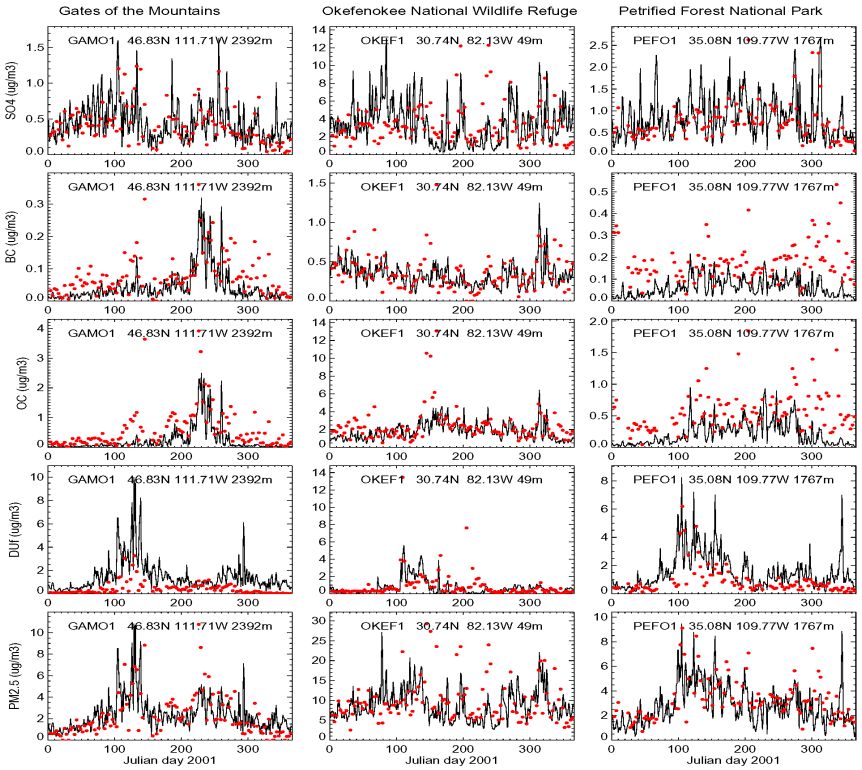

Figure 1. Daily average concentrations of sulfate (SO4), BC, OC, fine mode dust (DUf), and PM2.5 at 3 IMPROVE sites in 2001. Circles are IMPROVE data and lines are model results.

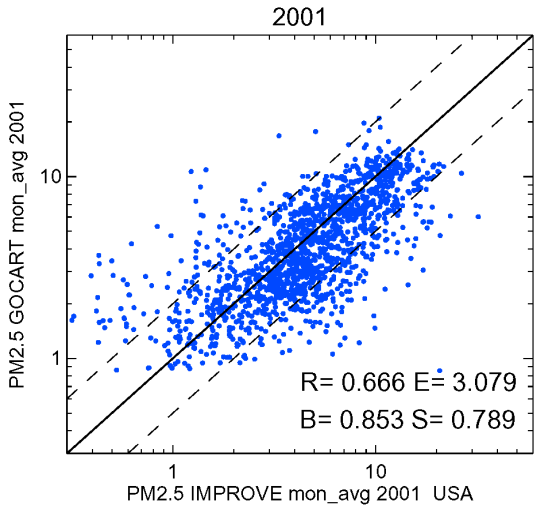

Figure 2. Comparison of monthly average PM2.5 concentrations in 2001 at 168 IMPROVE sites. R = correlation coefficient, B = mean bias, E = Root mean square error, S = skill score (see Chin et al. 2004 for definition).

Contribution of aerosols from different sources

In order to assess the contributions of aerosols over the U.S. from different source regions, we have run several model experiments in which a particular source or emission from a particular region is "zeroed out". Table 1 lists the model experiments.

Table 1. GOCART model experiments.

Exp.	Definition	Model setup
ALL	PM2.5 from all sources, including anthropogenic, biomass burning, and natural emissions	Standard model run
BKG	Background PM2.5 – from sources we cannot control, e.g. dust, volcanoes, trees, Long-range transport from other regions	Model run without North America anthropogenic emission (with biomass burning)
NAT	PM2.5 from natural sources only	Model run without all anthropogenic emission (with biomass burning)
BMB	PM2.5 from biomass burning	Differences between standard run and runs without biomass burning emissions
NAM ASA EUR	Sulfate aerosols from North America (NAM), Asia (ASA), or Europe (EUR) anthropogenic emissions	Differences between standard run with all sulfur sources and the run without anthropogenic emissions in a selected region
AFR ASA MDE	Dust aerosol from Africa (AFR), Asia (ASA), or Middle East (MDE)	Differences between standard run with all dust sources and the run without emissions in a selected region

Figure 3 demonstrates the model results of total PM2.5 and PM2.5 from background and natural sources for the four quadrants (NW, SW, NE, SE, divided at 100°W and 40°N) over the U.S. The model shows that in the western half of the U.S., about 70% PM2.5 is "background" with nearly 60% "natural" (e.g., dust, biogenic aerosols) on average. In the eastern half, local anthropogenic sources contribute to 60% of PM2.5 concentrations. Further analysis shows that the U.S. anthropogenic sources contributes to 70 – 90% of sulfate PM2.5 at the surface while dust from Asia could supply about half fine dust loading except in the SW quadrant where local deserts are the major source of dust (64%). The contribution of forest and agriculture fires varies significantly with location and seasons.

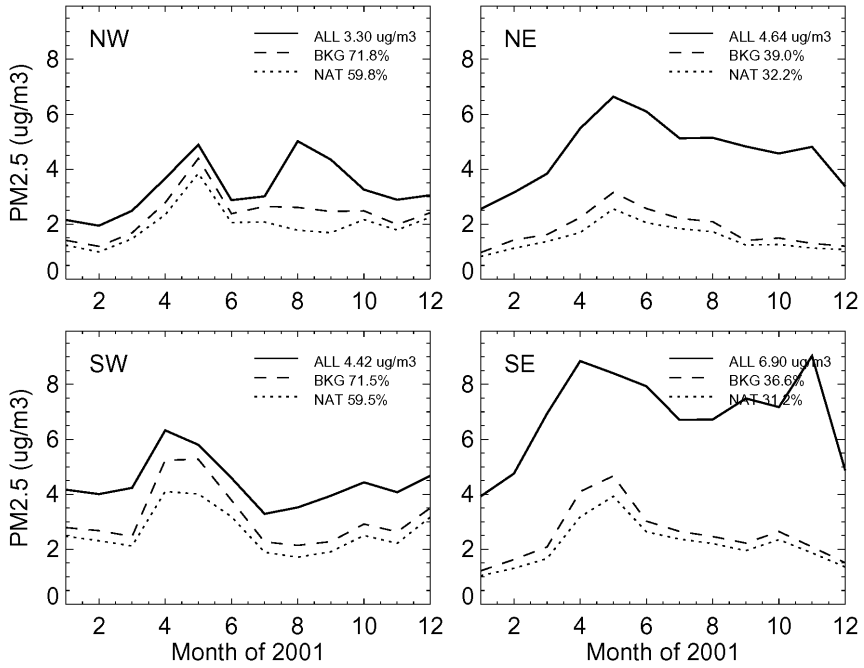

Figure 3. Monthly average surface layer PM2.5 concentrations in 2001 from the GOCART model (see Table 1 for experiment definition). Annual average concentrations and % contributions from different sources are shown. The difference between BKG and NAT is PM2.5 anthropogenic transport form other regions, and the difference between ALL and BKG is PM2.5 from North America own pollution sources (and biomass burning).

Relationship between AOT and PM2.5

As we mentioned in the "Introduction", it is important to understand the relationship between the column AOT and the surface PM2.5 in order to intelligently use the satellite data for air quality monitoring. A robust relationship between the column AOT and surface PM2.5 can be achieved only if a) the aerosol vertical profile maintains a relatively constant shape such that the change in total column AOT is nearly proportional to the change at the surface, and b) the aerosol composition stays the same such that the mass extinction efficiency, which converts aerosol mass to AOT depending on aerosol type, size, and relative humidity, remains more or less constant. Unfortunately, there is usually a significant heterogeneity in aerosol distributions and composition that make the AOT-PM2.5 relationship vary with location and season, as shown in a recent analysis using the AOT from the satellite data from the MODIS instrument and the surface PM2.5 measurements from more than 1000 sites over the U.S. (Engle-Cox et al., 2004). The concept is further illustrated in Figure 4 with the GOCART model results, which shows that the relationship between the AOT and surface PM2.5 varies from poor to excellent depending on location and time. In general, the correlation is better in summer/fall than in winter/spring and better over the eastern half of North America than over the western half.

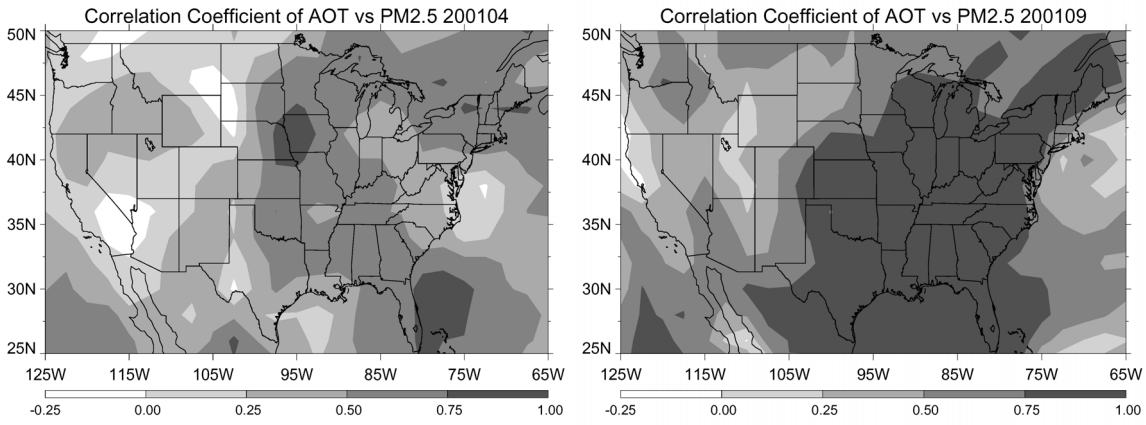

Figure 4. Correlation coefficients of daily AOT and PM2.5 for April (left panel) and September (right panel) 2001 from the GOCART model.

We further explain the cause of these different relationships by examining the PM2.5 composition and the aerosol vertical distributions at 2 sites, one located in the eastern U.S. (Ohio) and one in the west coast (Washington state). The spatial and temporal difference in the AOT-PM2.5 relationships can be explained by the differences in aerosol composition and vertical distributions. At the Ohio site, sulfate is the dominant aerosol type for both April and September and located mainly in the boundary layer. In contrast, aerosol compositions and vertical distributions are very different between April and September at the Washington State site located at the west coast. In spring, the site receives not only large amount of dust transported from Asia with plume extending to 5 km but also sea-salt aerosol from the Pacific. In late summer/early fall, on the other hand, the site is heavily influenced by biomass burning with carbonaceous aerosol dominating. Therefore, it is more difficult to quantitatively relate column AOT with surface PM2.5 in the western than in the eastern U.S. because of the larger variation of aerosol composition and vertical profiles in the west.

Conclusions

We have used the global model GOCART to investigate the relative contributions of regional anthropgenic emissions, forests and agriculture fires, and long-range transport of aerosols from other regions in the world to the surface PM2.5 concentrations at the U.S. that affect the air quality. The model study for 2001 indicates that in the eastern half of the U.S., regional anthropogenic emissions, natural aerosols, and long-range transport of anthropogenic aerosols from other regions contribute about 62%, 32%, and 6% to surface PM2.5, respectively, while in the western U.S. the percentages are 28%, 60%, and 12%. About 70 to 90% of sulfate aerosols over the U.S. are from the regional/local anthropogenic sources.

Satellite AOD data provide very helpful guidance for PM2.5 forecasts and monitoring. At the time and places when the aerosol composition and vertical distributions are stable, the column AOD can be quantitatively scaled to surface PM2.5 concentrations, such as in the eastern U.S., especially during warm months (summer/fall) that aerosols are predominantly of regional/local pollution origin and concentrated mostly within the boundary layer. However, in the western U.S. where aerosols are from multiple origins (local pollution, desert, long-range transport from Asia, biomass burning) direct link between AOD and PM2.5 is more difficult because the composition and vertical profiles have large seasonal variations.

To obtain a high quality PM2.5 prediction and assessment capability, a combination of large-scale model, satellite AOD data, and vertical profile information from either local lidar network or from future satellites (e.g. CALIPSO) is needed to quantitatively estimate the effects of long-range transport and separate boundary layer aerosols with aloft plumes.

Acknowledgments

We thank the IMRPOVE network scientists, site managers, and technicians for making the surface aerosol data available. This work is supported by NASA.

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