

## Managing Agricultural Emissions to the Atmosphere: State of the Science, Fate and Mitigation, and Identifying Research Gaps

S. R. Yates,\* L. L. McConnell, C. J. Hapeman, S. K. Papiernik, S. Gao, and S. L. Trabue

The impact of agriculture on regional air quality creates significant challenges to sustainability of food supplies and to the quality of national resources. Agricultural emissions to the atmosphere can lead to many nuisances, such as smog, haze, or offensive odors. They can also create more serious effects on human or environmental health, such as those posed by pesticides and other toxic industrial pollutants. It is recognized that deterioration of the atmosphere is undesirable, but the short- and long-term impacts of specific agricultural activities on air quality are not well known or understood. These concerns led to the organization of the 2009 American Chemical Society Symposium titled *Managing Agricultural Gas and Particle Emissions*. An outcome of this symposium is this special collection of 14 research papers focusing on various issues associated with production agriculture and its effect on air quality. Topics included emissions from animal feeding operations, odors, volatile organic compounds, pesticides, mitigation, modeling, and risk assessment. These papers provide new research insights, identify gaps in current knowledge, and recommend important future research directions. As the scientific community gains a better understanding of the relationships between anthropogenic activities and their effects on environmental systems, technological advances should enable a reduction in adverse consequences on the environment.

THE EFFICIENCY AND SUSTAINABILITY of agriculture production is highly dependent on interactions within the soil–water–atmosphere–plant–animal ecosystem. Soil, water, and the atmosphere provide a growth medium that enables sustained plant and animal production and play critical roles in exchanges of mass and energy and in recycling processes. Recent advances in agricultural practices have helped to maintain the world's supply of needed food and fiber; life expectancy has increased significantly and illnesses related to malnutrition have decreased over the last century (Borlaug, 2007). While modern agricultural systems have provided considerable benefits to the growing world population, these activities have also produced some undesirable environmental consequences including damage to soil, water, and atmospheric resources and deleterious effects on ecosystem and human health (Dockery and Pope, 1994; Dockery et al., 1996; Künzli et al., 2000; Schwartz, 2004).

The earth's atmosphere is highly dynamic, and emitted pollutants can travel globally depending on their volatility and reactivity. Predicting the fate of atmospheric pollutants and their effects on human and ecosystem health is highly complex and is dependent on meteorology, other pollutants/constituents present in the air, and the chemical, physical, and toxicological properties of the pollutant. Some of these effects, such as the formation of brown haze around cities in the summer due to photochemical smog, are readily observable, whereas other effects, such as chemicals that contribute to the destruction of the stratospheric ozone layer, are more subtle. Both animal husbandry operations (NRC, 2003; Faulkner and Shaw, 2008) and cropping systems (Pimentel et al., 1995; Krupa et al., 2008; Gomiero et al., 2008) can affect regional air quality. Constituents of concern include particulate matter, volatile organic and inorganic compounds, ozone, and pesticides. Furthermore, pesticide use (Pimentel et al., 1992; Pimentel, 1994; van den Berg et al., 1999) and the introduction of herbicide-tolerant crops can promote changes to agricultural environments that can translate into adverse effects on overall air quality (Graef, 2009). The effects of these emissions can be especially acute where urban and agricultural communities are in close proximity. In this situation, the agricultural

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\*Corresponding author (scott.yates@ars.usda.gov).

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5585 Guilford Rd., Madison, WI 53711 USA

S.R. Yates, USDA–ARS, U.S. Salinity Lab., 450 W. Big Springs Rd., Riverside, CA 92507; L.L. McConnell and C.J. Hapeman, USDA–ARS, Environmental Management & Byproduct Utilization Lab., Beltsville, MD; S.K. Papiernik, USDA–ARS, North Central Agricultural Research Lab., Brookings, SD; S. Gao, USDA–ARS, San Joaquin Valley Agricultural Sciences Center, Parlier, CA; S.L. Trabue, USDA–ARS, National Lab. for Agriculture and the Environment, Ames, IA. Assigned to Editor Dennis Corwin.

**Abbreviations:** AFO, animal feeding operation; NRC, National Research Council; PM, particulate matter; VOC, volatile organic compound.

system may create nuisance and offensive odors and/or toxic pesticide plumes or can contribute to near-surface ozone and aerosols, which can cause eye irritation or decreased lung function and lead to increased morbidity and mortality (Dockery and Pope, 1994; Takemoto et al., 2001; Bell et al., 2005). All of these result in increased tension between growers and nearby urban populations.

The importance of the interplay between agricultural systems and air quality is demonstrated by the intense and varied research activities, by the many review articles and commentaries that have been written, and by the numerous federal, state, and local programs, research efforts, funding opportunities, and regulatory activities. Review articles have considered greenhouse gas emissions (e.g., Visek, 1984; NRC, 2003; Johnson et al., 2007; Smith et al., 2007), the effect of animal feeding operations (AFOs) on regional air quality (e.g., NRC, 2003; Aneja et al., 2009), and the consequences of pesticide emissions, fate, and transport (e.g., Chesters et al., 1989; Pimentel, 1994; UNEP, 1998, 2006; van den Berg et al., 1999; Ruzo, 2006). Over the years, new regulations have been promulgated to control atmospheric emissions in hopes of preventing undesirable effects. Regulators rely on the observed effects, measured pollutant concentrations, landscape features, and modeling to predict pollutant fate and transport and to estimate potential risks. Early models were based on empirical observations, but as research efforts increase knowledge of emissions, fate, and atmospheric transport processes, a more deterministic approach becomes possible.

Research programs worldwide continue to recognize more fully the short- and long-term effects of agricultural activities on local and regional ecosystems and to investigate strategies to mitigate atmospheric deterioration due to agricultural emissions. In 2009, in Washington, DC, a symposium titled *Managing Agricultural Gas and Particle Emissions* was held at the 238th National Meeting of the American Chemical Society to examine the latest advances in measurement technologies, modeling tools, and mitigation strategies. The symposium objectives were (i) to review what is currently known about agricultural impacts on regional air quality and the environment, (ii) to present current research and identify research gaps related to agricultural impacts on air quality, and (iii) to present current information on methods to mitigate and/or manage emissions in an environmentally benign manner. The objectives of this paper are to provide an overview of the topic area, to introduce the papers that were presented in the symposium and that make up this special section of *Journal of Environmental Quality*, and to identify critical knowledge and research gaps that were identified by the participants.

## Measurement Methodologies

Monitoring gases or particulates in the atmosphere requires specialized sampling and analytical methodologies. A variety of gas sampling equipment, including sorbent materials, traps, evacuated canisters, and bags, are used in discrete sampling of gases, whereas size fractionation onto filters is used for discrete sampling of particulates (Waite et al., 2005). Gas and particulates are also measured continuously in real-time using both closed and open-path instruments that include Fourier trans-

form infrared spectroscopy (FTIR), selected ion flow tube-mass spectrometry (SIFT-MS), cavity ring down spectroscopy, and light detection and ranging (LIDAR) systems.

Researchers have also developed a variety of experimental methods to obtain emission values. Some of these include chamber methods (Hutchinson and Mosier, 1981), micrometeorological methods (Parmelee et al., 1972; Denmead et al., 1977; Wilson et al., 1982), inverse methods (Flesch et al., 1995; Johnson et al., 2010), and soil mass balance methods (Yates et al., 1996b; McIsaac et al., 2002; Weldon and Hornbuckle, 2006). These methodologies have different sensitivities and require different levels of experimental and theoretical sophistication. In some cases, the methodologies may lead to biased results or may suffer from theoretical constraints that could lead to erroneous or uncertain emission values.

## Measurement Uncertainty

Many soil, atmospheric, and cultural factors affect emission processes and make obtaining accurate emission estimates difficult. Furthermore, the relative importance of these factors may change based on regional or local conditions. When pollutant concentrations are very low, the uncertainty in flux calculations can be quite high. In their study, Myles et al. (2011) measured the depositional velocity of several gases over a large unfertilized field, comparing values based on profile flux measurements with estimates obtained using a resistance analogy. Observed discrepancies between the two approaches were attributed to errors associated with measuring low concentrations in the atmosphere and the formation of dew on the canopy, which can increase deposition rates. This study provides important information to assist researchers in developing more accurate and robust methodology to measure atmospheric emissions and deposition, particularly for nutrient compounds. This research also clearly shows a need to obtain a more detailed understanding of the factors affecting emission and deposition processes.

## Improving Sampling Efficiency

Another issue in the measurement of gas flux in field settings is the high cost and complexity of conducting field experiments. This has led researchers to investigate ways to optimize the sampling design and sampling protocol to increase experimental information while reducing field sampling costs. A corollary to improving efficiency is ensuring that the experimental uncertainty is within acceptable limits. Any effort to reduce sampling costs must be weighed by the effect on experimental error and uncertainty. Assuming that the methodology provides sufficiently precise and accurate results, any improvement in sampling efficiency allows for more monitoring to be completed within a research budget. The research effort described in the paper by Sullivan and Ajwa (2011) addresses this issue. Sullivan and Ajwa describe research to measure the flux of trace gases over a landscape using the integrated horizontal flux technique using the results from five field experiments. This paper considers a variety of issues and complicating factors that can affect the outcome from field-scale experimentation. The authors provide information on the uncertainty and the precision of the integrated horizontal flux method for determining emission rates.

The reports by Myles et al. (2011) and Sullivan and Ajwa (2011) also support results by other researchers that the overall uncertainty of field-scale flux measurements is commonly in the 20 to 50% range (Businger, 1986; Wilson and Shum, 1992; Majewski, 1997; Liu and Foken, 2001). It is important to acknowledge uncertainty in the methodology used to determine fluxes as the uncertainty places limits on the achievable accuracy of measured emissions. Significant improvements in theory and/or equipment/sampling methodology would be required to achieve a significant increase in accuracy. This is an important area in need of further research.

### **Agricultural Film Permeability**

Plastic films have been used for decades to reduce diffusion losses and gaseous transport through food packaging (Johansson and Luefven, 1994; Piringner et al., 1998), to contain pesticides in soil (Wang et al., 1997b; Wang and Yates, 1999; Gao and Trout, 2007), and to mitigate ammonia emissions (Scotford and Williams, 2001; Sommer, 2001; Portejoie et al., 2003). The properties of a membrane and, in particular, the rate of chemical diffusion through the membrane are of critical importance in making efficient and cost-effective use of agricultural resources and efforts to protect the environment.

Several published methods are available to determine film diffusion resistance (i.e., mass transfer coefficient). These include methods based on flow-through designs (Kolbezen and Abu-El-Haj, 1977; Wang et al., 1999) and methods that utilize static cell devices (ASTM, 1982; Papiernik et al., 2001, 2002; Austerweil et al., 2006). Current standardized methods produce variable (ASTM, 1982) and often insensitive (Gamliel et al., 1998) measures of diffusion through membranes.

With the introduction of low-permeability films (i.e., virtually impermeable films) and increasing regulation of soil fumigation has come an increasing need for simple, accurate, and dependable methods to measure the permeability of a film for use in soil fumigation. Papiernik et al. (2011) describe a new approach for standardizing film permeability measurements that is based on measuring changes in gas concentration across a film sample. Using this approach, mass transfer coefficients (or equivalently film resistance,  $R$ , values) can be determined, enabling improved classification of films and accurate prediction of fumigant emissions through the film.

### **Agricultural Air Quality and Animal Feeding Operations**

Animal feeding operations can have a significant effect on regional air quality. The primary pollutants of concern include volatile organic compounds (VOCs), particulate matter (PM; i.e., coarse particulates,  $PM_{10}$  and fine particulates,  $PM_{2.5}$ ), and ammonia ( $NH_3$ ), which also serves as a secondary particulate matter (NRC, 2003). Other problematic emissions include methane ( $CH_4$ ), nitrous oxide ( $N_2O$ ), various nitrogen oxides ( $NO_x$ ), and hydrogen sulfide ( $H_2S$ ). Numerous compounds with strong and objectionable odors have also been identified as emanating from AFOs. While odors are usually considered nuisances, they often lead to complaints from nearby residential communities and can eventually lead to reduced agricultural activities.

The USEPA recently estimated ammonia emissions from confined and rangeland animal feeding operations in the United States for the years 2002, 2010, 2015, 2020, and 2030 (Fig. 1). Ammonia emissions are projected to be nearly 2400 Gg per year nationally by 2030, an increase of 12% over 2010. Dairy and beef production is the largest contributor to the national ammonia inventory, with approximately 50% of the total emissions (Fig. 1; NRC, 2003; USEPA, 2004).

Agriculture is also a significant source of  $CH_4$  and  $N_2O$  to the atmosphere. On a global basis, the estimated agricultural emissions of  $CH_4$  and  $N_2O$  are about 47 and 58% of the total anthropogenic sources (Smith et al., 2007). For  $N_2O$ , emissions are primarily associated with N fertilization and application of animal manure. In regions associated with large livestock numbers, high levels of  $CH_4$  occur due to enteric fermentation (USEPA, 2006). Large emissions of  $CH_4$  are also associated with rice production.

Agricultural emissions of  $CH_4$  and  $N_2O$  are likely to increase significantly by 2030 (e.g., 30% or more) due to a combination of increased fertilizer use, increased livestock and rice production, and increased land application of animal manure (Smith et al., 2007). The USEPA (2006) estimated that  $N_2O$  emission will increase from approximately 8 Tg in 1990 to over 11 Tg by 2020, with emissions from soil representing the largest source (Fig. 2). Future estimates have relatively high levels of uncertainty because the effects of potential mitigation methodologies to decrease emissions remain largely unknown.

Sustaining agricultural production is a critical component to global food security. Without well-functioning agricultural systems, global food supplies will be reduced, potentially leading to widespread famine. To maintain agricultural production, methodology is needed to mitigate deterioration of soil, water, and air resources on which agricultural systems depend. There will be a greater need to understand the interrelationships between soil–water–air systems to determine the effect that changes in management practices (often driven by regulations) might have across environmental compartments/sectors. For example, the National Research Council (NRC, 2003) suggests that regulations for AFOs aimed at improving water quality could lead to increased emissions to the atmosphere. Clearly, sound science is needed to develop regulations that protect all environmental compartments while maintaining agricultural production.

Furthermore, because of the complexity of interactions between agriculture and the environment, it is likely that both research and regulatory models will be needed to integrate processes, interactions, and effects across landscapes. The use of simple approaches, such as basing emission inventories on per animal emission factors, has a number of weaknesses and is unsatisfactory for estimating annual loading or the temporal behavior of the emissions (NRC, 2003). It has been suggested that regulators make use of a more comprehensive process-based modeling approach that is constrained by mass inputs. This would also alleviate unrealistic outcomes where simulated total emissions exceed input levels (NRC, 2003).

### **Ammonia and Trace Gas Emissions**

In a recent report, the NRC (2003) indicated the need for more on-farm emission estimates as a means for determining

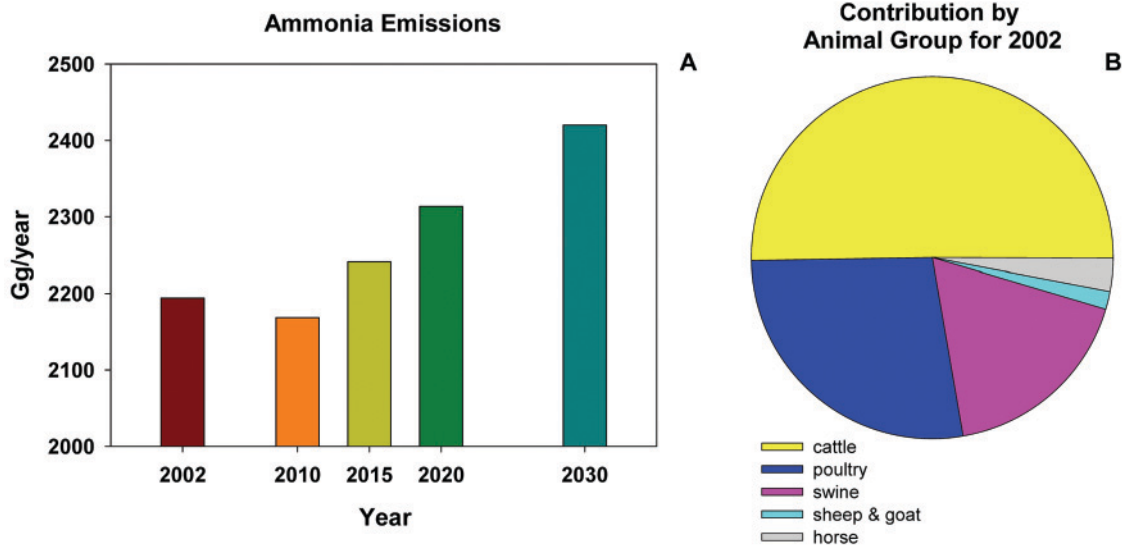


Fig. 1. (a) Estimated ammonia emissions (Gg/yr) from 2002 until 2030 from animal feeding operations; the data include cattle, swine, sheep, goat, and poultry. (b) The contribution (%) from each animal group using data from 2002. Adapted from USEPA (2004).

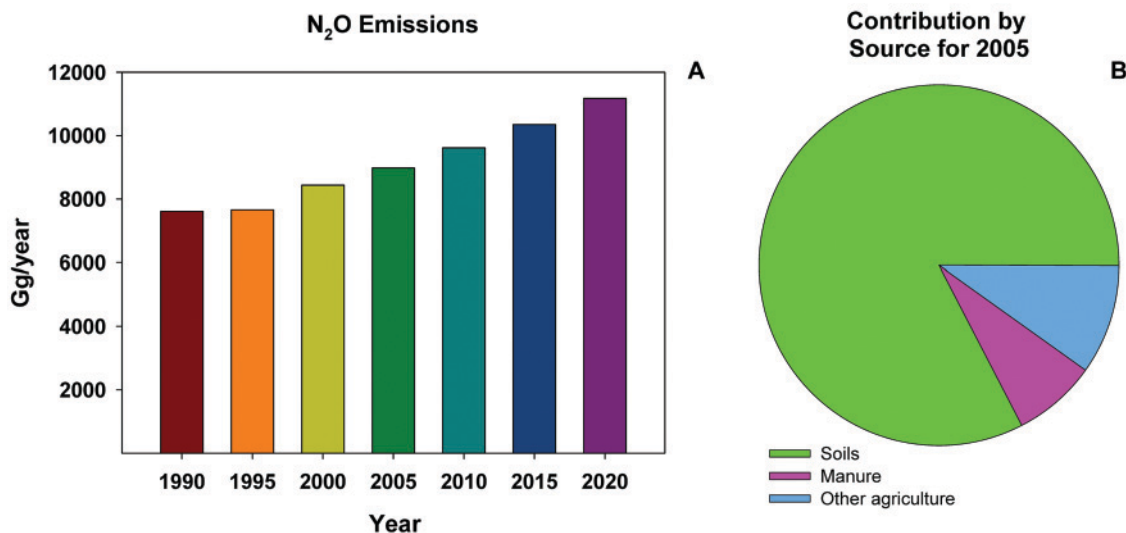


Fig. 2. (a) Estimated N<sub>2</sub>O emissions (Gg/yr) from 1990 until 2020 from agricultural sources. (b) The contribution (%) from each source group using data from 2005. Adapted from USEPA (2006).

more accurate annual inventories. To accomplish this, methods are needed to gather accurate information regarding on-farm emission rates that take into account both diurnal and seasonal variations under different production scenarios and climatic regions. In their paper, Leytem et al. (2011) provide details of a study to determine NH<sub>3</sub>, CH<sub>4</sub>, CO<sub>2</sub>, and N<sub>2</sub>O emission rates from open lots, wastewater pond, and compost source areas over a year time span. Their results provide important information on emissions from an open-lot dairy located in southern Idaho topographic and climatic settings. They found that open-lot areas were the largest sources of these trace gases, with temporal variation in emissions from each source area. They also indicate that when regulating emissions, many aspects of the problem should be considered. For example, while CO<sub>2</sub> emission by cows in open lots may be higher than pasture-based systems, when put on a production basis (i.e., milk produced), the CO<sub>2</sub> per quantity milk is lower. This information could help lead to informed regulatory decisions and, through

optimization, may lead to approaches to minimize emissions while maximizing the agricultural products.

Agricultural emissions from poultry facilities in Arkansas were studied by Moore et al. (2011), who provide in their paper the first measurements in the USA of NH<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub>, and CO<sub>2</sub> emissions during rearing, from litter during storage and after land application. They used this information with other N inputs and outputs to obtain an N mass balance approaching 100% for the poultry operation. The authors also found that NH<sub>3</sub> emissions can be reduced by knifing the litter into soil. In this study, approximately 50 g of NH<sub>3</sub> was produced per marketable bird, considering emissions in the poultry house and during litter disposal.

### Odors

Animal and/or municipal waste offer a valuable resource to improve soil tilth and provide nutrients for plant growth. A negative side effect of using this material is the emission



of compounds with offensive odors. Reducing emission of offensive compounds would make this material a more attractive byproduct and would also provide a needed avenue for recycling potentially useful material. However, reduction of odors first requires careful study and identification of the offending chemicals, including the development of new sampling methods (Trabue et al., 2008; Zhang et al., 2010) and relating animal production to release of odorants (Blanes-Vidal et al., 2009).

Laor et al. (2011) provide new information on a range of chemicals emitted from biosolids. They found that several classes of sulfur-containing compounds make up the most offensive odors and suggest their use as chemical indicators of odor from biosolids. They also found that alkaline stabilization tended to lead to high emissions of N compounds and did not reduce odor. In their study, anaerobic conditions in soil led to odoriferous compounds being emitted from soil weeks after application.

One approach being investigated for mitigating odors is modification of animal diet. To test this approach, Woodbury et al. (2011) utilized electromagnetic sensing equipment to study the spatial distribution of volatile fatty acids in a feedlot. This equipment was used to identify soil sampling positions in pens with cattle fed different diets. Soil samples were incubated and the fermentation products were analyzed to categorize the structure of the volatile fatty acids. These researchers determined that diet is related to the amount and offensiveness of volatile fatty acid odors emitted from a feed lot. This approach may lead to future mitigation of odiferous emissions from animal waste materials.

## Particulate Matter

Agricultural operations can contribute significantly to the emissions of PM to the atmosphere. Exposure to PM has serious health consequences and is linked to increased heart and lung disease and to premature mortality (Samet et al., 2000). Particles with diameters of 10  $\mu\text{m}$  or less can penetrate the lungs deeply, and very small particles can pass through the lungs and affect other organs. Exposure to PM may even have the potential to affect nonrespiratory systems such as the central nervous system (Ngo et al., 2010).

Airborne particles with aerodynamic diameters that are equal to 10  $\mu\text{m}$  and 2.5  $\mu\text{m}$  or less are referred to as  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , respectively. The particle-size fraction less than 2.5  $\mu\text{m}$  has received much more regulatory attention and associated research due to its potential to cause severe health effects compared with larger-sized particles. In response to elevated risk, the USEPA (2009) revised the particulate air quality standards in 2006 and lowered the 24-h average  $\text{PM}_{2.5}$  level from 65 to 35  $\mu\text{g m}^{-3}$ , while maintaining the 24-h average  $\text{PM}_{10}$  level at 150  $\mu\text{g m}^{-3}$ .

Particulates resulting from AFOs and agricultural machinery used to prepare land for planting and harvesting are significant sources of PM to the atmosphere. Emission of PM from land preparation exhibits a seasonal pattern that varies depending on field activities associated with specific crops, and emissions related to AFOs depend on the structure of the facility and the activity of animals (Cambra-López et al., 2010; McGinn et al., 2010). Emissions of PM from agricultural operations were

estimated to be approximately 9% of total suspended particles, 11% of  $\text{PM}_{10}$ , and 11% of  $\text{PM}_{2.5}$  in Canada in 2006; most of the PM emissions were from wind erosion and land preparation (Pattey et al., 2010).

Physical characteristics of emission sources, particle-size distributions of the PM emitted, and natural dispersion of emissions into the environment cause difficulties in quantifying PM emissions from agricultural operations (Wanjura et al., 2009). Because uncertainties in emissions are considered substantial, extensive research has been done on correcting and improving existing methodologies as well as developing new methods and information (Wang et al., 2005; Waite et al., 2005; Simon et al., 2008; Wanjura et al., 2009). There continues to be a significant research need for accurate methods to quantify particulate emissions from agricultural operations.

Many studies have investigated PM emission reduction strategies involving agronomic practices or modification of machinery operations, such as reduced-pass almond sweeping (Goodrich et al., 2009). Conservation tillage, no-till practices, and a decrease in the area of summer-fallow land were attributed to the reduction of PM emissions in Canada from 1981 to 2006 (Pattey et al., 2010). However, modifying machinery to capture PM before it enters the atmosphere can be costly and may result in the emission of other pollutants and greenhouse gases (Funk, 2010). It is clear that much more research is needed for methods to accurately estimate emissions from agricultural sources, for determination of particle size, composition, and distribution, and for the development of methods to reduce emissions of PM.

## Ozone Formation from Agricultural Emissions

Ground-level ozone is both a public and an environmental health hazard. Ozone is a main component of photochemical smog, and exposure to ozone can reduce lung function and produce respiratory inflammation in humans. The elderly and children are especially susceptible to respiratory inflammation (Brunekreef and Holgate, 2002). High ozone levels are also associated with higher rates of mortality and morbidity and lead to increased health-care costs (Bell et al., 2005). High ozone levels can also create economic loss by damaging crops and reducing productivity (Fangmeier et al., 1994).

Ground-level ozone is a severe problem in the San Joaquin Valley of California and in numerous urban areas throughout the United States (Fig. 3). Ozone is formed in the atmosphere by the reaction of VOCs and  $\text{NO}_x$  in the presence of sunlight. The highest ozone levels are typically found in suburban areas because of the transport of precursor emissions from the urban center, and local topographic effects can exacerbate ozone levels. From 1980 to 2009, summer emissions of  $\text{NO}_x$  decreased by nearly 50% from 30 to 15 million metric tons per year (Table 1). In 2005, the principal sources of  $\text{NO}_x$  emissions (Table 2) were found to be on-road vehicles (36%), off-road sources (23%), and the generation of electricity (21%). Furthermore, VOC emissions in the United States decreased significantly from over 33 million metric tons in 1980 to about 14 million metric tons in 2009 (Table 1).

In areas heavily influenced by agriculture, pesticides can become a significant contributor to VOC emissions. In 2004,

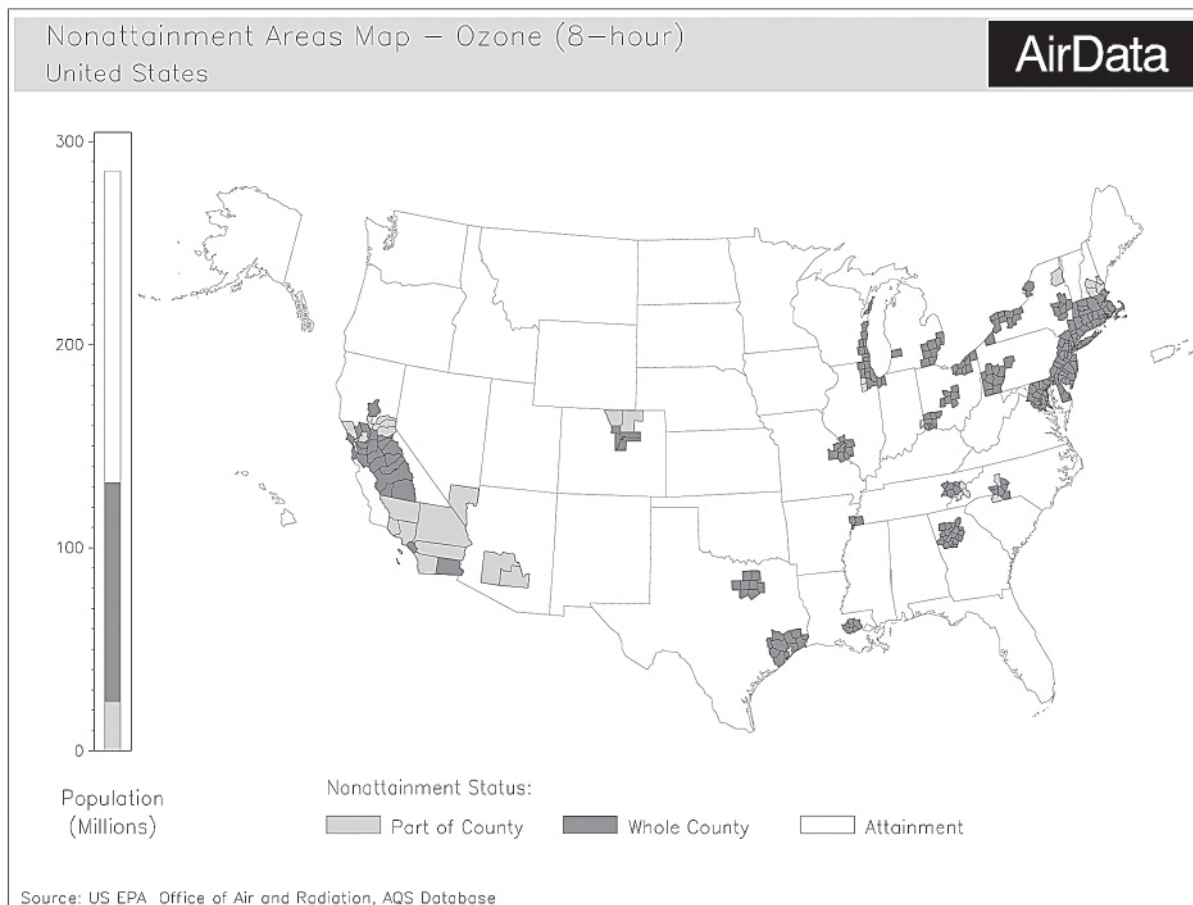


Fig. 3. Map of nonattainment designations for 8-h ozone in the United States as of 16 Dec. 2008. Source: USEPA (2011c).

for example, pesticides contributed approximately 6.3% to the total VOC emissions in the San Joaquin Valley (Table 3). Pesticide use in these areas may have a significant effect on VOC levels in the atmosphere and has led state regulators to develop plans to reduce pesticide emissions.

The USEPA has established federal 8-h ozone standards that require state regulators to develop and submit state implementation plans (SIPs) to meet the air quality standards. Generally, this involves the need for reductions in emissions from many sources, including pesticides. This has led to recognition that more research is needed to determine the impact of pesticide emissions on ozone formation and to develop methods to reduce emissions. In regions with significant agricultural production, reducing VOC emissions from solvents and additives in pesticide formulations and from the use of soil fumigants will likely be targeted in an effort to comply with national ambient air quality standards.

Inventory reduction targets for pesticide emissions have been set by the California Department of Pesticide Regulation (CDPR, 2009). These inventories were developed by assuming complete loss of the VOC portion of semivolatile pesticide formulations. This approach leads to an overestimation of soil pesticide emissions because abiotic and biotic degradation and irreversible sorption, which can reduce soil concentrations, are not considered. Additional research is needed to develop more accurate methods to determine emission rates for these pesticides.

For soil fumigants, a relatively large number of field experiments have been conducted to measure emission rates, providing regulators with sufficient data to develop an approach to adjust total fumigant emissions by associating emission factors to particular fumigant and application scenarios. Although this is a significant improvement over semivolatile pesticides, further research is needed to determine how soil, atmospheric, and fumigant management conditions affect the emission processes so that these factors can be addressed in the regulations.

Research is needed to accurately determine VOC emissions from pesticide application and from soil fumigation and to develop methods to reduce emissions to low levels. Failure to do so may cause agricultural producers to face potentially restrictive control strategies, which may cause a reduction in profit or force farmers to cease food production.

### Direct Measurement of Ozone

A complicating factor in meeting air quality standards is determining the effect of VOC emissions on the production of O<sub>3</sub>. With a few exceptions (Howard et al., 2008), there is a lack of information on the reactivity of many VOCs and participating species, which leads to highly uncertain estimates of the effect of VOC on O<sub>3</sub> production. To this end, Kumar et al. (2011) determined the constituents of VOCs in the air surrounding fields before and after application of a pesticide. Using portable smog chambers, they found that this VOC profile elevated O<sub>3</sub> formation at two of four orchard sites for 1 to 2 d following

**Table 1. Estimated pollutant emissions† in the United States (millions of metric tons/yr).**

Pollutant	Year						
	1980	1985	1990	1995	2000	2005	2009
Carbon monoxide	196	187	159	132	112	103	77
Sulfur dioxide	29	25	25	21	18	17	10
Lead	0.082	0.025	0.006	0.004	0.003	0.002	0.002
Nitrogen oxides	30	29	28	28	24	21	15
Volatile organic compounds	33	30	25	24	19	20	14
Particulate matter (PM)							
PM <sub>10</sub> ‡	7	4	3	3	2	2	1
PM <sub>2.5</sub> ‡	NA§	NA	2	2	2	1	1
Totals	294	276	240	208	175	162	118

† Fires and dust excluded. Adapted from USEPA (2011b).

‡ PM<sub>10</sub> and PM<sub>2.5</sub> refer to particles with aerodynamic diameters that are equal to 10 μm and 2.5 μm or less, respectively.

§ NA, not available.

**Table 2. Estimated pollutant emissions contribution by source† in 2005 (%).**

Pollutant	Electricity generation	Fertilizer & livestock	Fires & combustion‡	Industrial processes	Misc.	Nonroad equipment	On-road vehicles	Solvent use	Waste disposal	Road dust
Carbon monoxide	0.8	–	6.8	2.7	0	25	58.8	0	2.4	–
Lead	2.3	0	7.9	41.2	0.4	41.9	–	1.2	5	–
Sulfur dioxide	72.9	–	15.5	7.8	0	2.5	1	0	0.2	–
Ammonia	0.6	83.3	2.2	5.2	0	0.1	7.6	0	0.8	–
Nitrogen oxides	20.7	0	13.5	6.4	0	22.8	35.5	0	0.8	–
PM <sub>10</sub> §	3.1	0.1	4.4	6.2	27.8	1.4	1	0	1.5	52.5
PM <sub>2.5</sub> §	11.5	0	14	12.1	17	6	3	0.2	6.2	21.5
Volatile organic compounds	0.3	0.3	5.2	10.3	7.5	17.8	25.8	26.6	2.9	0

† Adapted from USEPA (2011a).

‡ Includes residential wood burning, fossil fuel combustion and fires.

§ PM<sub>10</sub> and PM<sub>2.5</sub> refer to particles with aerodynamic diameters that are equal to 10 μm and 2.5 μm or less, respectively.

pesticide application. Direct measurements of O<sub>3</sub> production are very important to ensure protection of environmental resources and to develop appropriate regulatory programs that do not needlessly impact farming operations or that are costly but ineffective. Continuing this research will also lead to a better understanding of the relationships of pesticide VOC emissions and ozone production.

Although direct measurement of O<sub>3</sub> from changes in VOC concentrations provides very useful information, it is impractical to frequently obtain this information over large regions via direct measurement. Furthermore, it is difficult to obtain information that would be useful in developing risk assessments or mitigation strategies or determining regional uncertainty in concentration.

## Emissions of Pesticides

Pesticides are a component of total VOC emissions, but they also pose an additional hazard when entering the atmosphere. For highly toxic pesticides, exposure to vapors can be a significant health issue. For soil fumigants and other pesticides, the short-term peak emission events are of regulatory concern, and bystander exposure risk depends on emission rates, atmospheric transport conditions, and proximity to the application site.

Many soil, pesticide management, atmospheric conditions, and other factors influence the rate of emissions from soil.

Emissions of pesticides from soil have received considerable study since the mid-1990s, and research has shown that large emissions fractions are possible for pesticides with a wide range of vapor pressures (e.g., Glotfelty et al., 1984; Majewski et al., 1993; Yagi et al., 1995; Yates et al., 1996a; Prueger et al., 2005; Yates, 2006; Gish et al., 2011).

It is extremely rare for researchers to conduct multiple volatilization experiments at the same location. In their paper, Gish et al. (2011) describe an 8-yr study in which they conducted repeated emission experiments at the same site without any changes in field management or pesticide application methodology. This series of experiments provides a rare look at the temporal variability of the total and the short-term emission patterns and provides unique information that can be used to understand the effect of various environmental processes

**Table 3. Major sources of volatile organic carbon in San Joaquin Valley 2004 (CDPR, 2006).**

Category	Emissions
	%
Livestock waste (dairy cattle)	9.6
Light- and medium-duty trucks	9.1
Light-duty passenger cars	8.3
Prescribed burning	7.5
Oil And gas production	7.4
Pesticides	6.3
Consumer products	6.2

on pesticide fate and transport. The study also found that total volatilization far exceeded runoff losses for the studied herbicides. This research provides a unique data, set and further research will provide a unified framework to explain the observed year-to-year differences.

## Soil Fumigants

Soil fumigant chemicals, which are VOCs, are effective at controlling a variety of plant pests, including nematodes, plant pathogens, weeds, and insects (Noling and Becker, 1994; UNEP, 1998), and help ensure the economic viability of many important crops, including strawberry (*Fragaria* L.), tomato (*Solanum lycopersicum* L.), pepper (*Capsicum* L.), eggplant (*Solanum melongena* L.), tobacco (*Nicotiana tabacum* L.), ornamentals, nursery stock, vines, and turf (Ferguson and Padula, 1994). However, recent research has clearly demonstrated that fumigant emissions commonly exceed 25% of application rates (Yagi et al., 1995; Yates et al., 1996a; Sullivan et al., 2004; Gao and Trout, 2007), and methods to reduce emissions are needed (Yates et al., 2002). The high emission rate for soil fumigants is generally due to their relatively low soil adsorption and high vapor pressures and is an especially acute challenge in agricultural regions with urban encroachment because of the high potential for human exposure. Delineating the fate and transport and the extent of the affected areas is frequently accomplished using mathematical models, the output of which are often used by regulators. Simulation models play an important role in better understanding the fate and transport process, in determining emission fractions, and in the development of improved pest management practices.

In their study, Wang et al. (2011) used two fumigant fate and transport models to demonstrate that total emissions of the fumigant chloropicrin could be accurately simulated using either complex (i.e., numerical) or simplified (i.e., analytical) solution techniques. They found their simulations of shank-applied chloropicrin were accurate when the initial chemical distribution was uniform across the shank fracture; simulating the initial chemical distribution as a concentrated point source underpredicted emissions. This research should help regulators develop tools for determining emissions factors that are more specific to actual conditions and that account for local effects.

As researchers develop the ability to accurately simulate fumigant emissions, it becomes possible to study and integrate the effects of soil fumigation across landscapes. Cryer and van Wesenbeeck (2011) used a stochastic framework to investigate the effect of fumigant management on regional air quality. They conducted a study that coupled field observations, a soil emission model, and a regulatory air dispersion transport model to obtain information on chemical distribution, uncertainty, and risk. A key aspect of this research is that the results are linked to experimental data, which helps to anchor the results to real-world outcomes and ensure the simulations and outcomes are realistic. From their results, a variety of management options to reduce emissions can be explored and the effects at the regional scale can be estimated.

Incorporating a stochastic determination of input parameters allows an evaluation of the short- and long-term risk of exposure to soil fumigants. van Wesenbeeck et al. (2011) describe an approach to stochastically link field-scale fumigant

emission rates to an atmospheric dispersion model to determine the risk of chronic exposure to a fumigant in coastal and interior townships in California. This study shows how changes in fumigant use in a township potentially affect a population's lifetime exposure. Further research is needed to develop and test improved stochastic and deterministic fate and transport models and to provide regulators with a variety of valuable tools that can be used to protect human and ecosystem health.

## Mitigating Agricultural Emissions

Development of pest-control technologies that reduce reliance on chemicals or minimize pesticide emissions to very low levels is needed to decrease the negative consequences on environmental and human health associated with pesticide use. Several nonchemical control methodologies have been proposed for pest control, including steam sterilization (Awuah and Lorbeer, 1991; Luvisi et al., 2006), solarization (Katan, 1981; Hartz et al., 1993; Chellemi et al., 1997; Gallo et al., 2007), biocontrol (Jayaraj and Radhakrishnan, 2008), natural products (Matthiessen and Kirkegaard, 2006; Coelho et al., 1999), and low-input or organic farming approaches (Lebbink et al., 1994; Reganold et al., 2001), to mention a few. Nonchemical methods offer the potential to grow crops without the use of pesticides, thereby eliminating the impact on human and environmental health. To date, however, these methodologies have not been widely adopted.

In addition to reducing atmospheric emissions, new emission-reduction strategies need to minimize fumigant movement to surface or groundwaters, where they can also affect aquatic ecosystems (Merriman et al., 1991; Obreza and Onterman, 1991; Yon et al., 1991; Schneider et al., 1995).

Recent studies have shown that fumigant emissions may be significantly attenuated through the use of plastic surface barriers (Majewski et al., 1995; Gan et al., 1997; Wang et al., 1997b), surface water seals (Jin and Jury, 1995; Gan et al., 1996; Wang et al., 1997a; Gao and Trout, 2007; Gao et al., 2008; Yates et al., 2008), deep injection (Reible, 1994; Majewski et al., 1995; Yates et al., 1997; Wang et al., 1997a), fertilizer amendments (Gan et al., 2000; Zheng et al., 2004; McDonald et al., 2008), organic amendments (Smelt et al., 1989a,b; Gan et al., 1998; Dungan et al., 2005; McDonald et al., 2008), combining organic amendments with plastic films or surface water seals (Gao et al., 2009), and application of fumigants in drip irrigation systems (Schneider et al., 1995; Sullivan et al., 2004). Although much experimental information has been obtained, the results are sometimes contradictory and significant research gaps remain. For example, some laboratory and field-plot studies showed that increasing the organic matter content at the soil surface reduces 1,3-dichloropropene emissions (Gan et al., 1998; Dungan et al., 2005; McDonald et al., 2008), whereas other studies found that application of manure alone was ineffective (Gao et al., 2009, 2011). Further research is needed to develop a complete understanding of relevant mechanisms and processes affecting pesticide fate and transport, and newly developed emission-reduction methods need to be tested in production systems.

Although experimental results for the effect of organic amendments on fumigant emissions appear contradictory, increasing the soil organic matter content should increase



chemical degradation and lead to a reduction in fumigant emissions. To test this, Yates et al. (2011) conducted a large-scale field study to evaluate the use of composted municipal green waste amendments to reduce emissions of the fumigant 1,3-dichloropropene. The results indicate that soil incorporation of composted municipal green-waste reduced emissions by as much as 75 to 90% over conventional fumigation practices. Although this experiment was intended to isolate the effect of organic material on emissions, the results also suggest that other factors may be important in reducing emissions rates. For example, Reichman et al. (personal communication, 2011) showed that reductions in soil water content can lead to increases in vapor sorption, which can significantly suppress emissions. More research is needed to determine the relative importance of various factors that affect emissions.

Although many technologies have been proposed to reduce fumigant emissions, relatively few experimental studies have been conducted under typical agricultural conditions that produced side-by-side comparisons. In a series of field-plot experiments, Gao et al. (2011) compared several promising emission reduction approaches. The authors conducted the fumigant application using standard industry equipment and procedures to produce results that are compatible with large field-scale operations. This research confirms that using low-permeability plastic films offers the most effective and reliable approach to reduce fumigant emissions; their use would require minor modification of typical fumigation practices. The authors also found that the glue joint between film sections did not compromise the barrier integrity. Results such as those presented in Gao et al. (2011) are critical for the effective implementation of practices to mitigate VOC emissions and provide a demonstration that fumigant emissions can be reduced using relatively low-cost and simple-to-use techniques. However, further research is needed to demonstrate and test the reliability of mitigation methodology across landscapes and seasons, which, to date, remains a challenge.

## Concluding Remarks

A balance must be found between the benefits of food production and allowable ecosystem alteration. Although emission reduction may be a key factor in reducing agriculture's influence on the environment and human health, ultimately, uncontrolled population growth is the dynamic that leads to many environmental problems. As long as populations grow, there will be increased need for food production and increasing demands on natural resources. Even with new technology to reduce agriculture's footprint on the environment, increasing population would ultimately overwhelm any new technological advance and lead to a worsening environmental and public health.

In the future, the research community will be challenged with finding new solutions and developing improved technology that leads to more efficient agricultural production and reduced impact on environmental quality.

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