

Effects of Agriculture upon the Air Quality and Climate: Research, Policy, and Regulations

VINEY P. ANEJA*

Department of Marine, Earth, and Atmospheric Sciences,
North Carolina State University,
Raleigh, North Carolina 27695-8208

WILLIAM H. SCHLESINGER

Cary Institute of Ecosystem Studies,
Millbrook, New York 12545

JAN WILLEM ERISMAN

Energy Research Centre of The Netherlands,
P.O. Box 1, 1755 ZG Petten, The Netherlands, and
Department of Earth Sciences, VU University of Amsterdam,
1081 HV Amsterdam, The Netherlands

Received August 29, 2008. Revised manuscript received
March 13, 2009. Accepted April 8, 2009.

Scientific assessments of agricultural air quality, including estimates of emissions and potential sequestration of greenhouse gases, are an important emerging area of environmental science that offers significant challenges to policy and regulatory authorities. Improvements are needed in measurements, modeling, emission controls, and farm operation management. Controlling emissions of gases and particulate matter from agriculture is notoriously difficult as this sector affects the most basic need of humans, i.e., food. Current policies combine an inadequate science covering a very disparate range of activities in a complex industry with social and political overlays. Moreover, agricultural emissions derive from both area and point sources. In the United States, agricultural emissions play an important role in several atmospherically mediated processes of environmental and public health concerns. These atmospheric processes affect local and regional environmental quality, including odor, particulate matter (PM) exposure, eutrophication, acidification, exposure to toxics, climate, and pathogens. Agricultural emissions also contribute to the global problems caused by greenhouse gas emissions. Agricultural emissions are variable in space and time and in how they interact within the various processes and media affected. Most important in the U.S. are ammonia (where agriculture accounts for ~90% of total emissions), reduced sulfur (unquantified), PM_{2.5} (~16%), PM₁₀ (~18%), methane (~29%), nitrous oxide (~72%), and odor and emissions of pathogens (both unquantified). Agriculture also consumes fossil fuels for fertilizer production and farm operations, thus emitting carbon dioxide (CO₂), oxides of nitrogen (NO_x), sulfur oxides (SO_x), and particulates. Current research priorities include the quantification of point and nonpoint sources, the biosphere–atmosphere exchange of ammonia, reduced sulfur compounds, volatile organic compounds, greenhouse gases, odor and pathogens,

the quantification of landscape processes, and the primary and secondary emissions of PM. Given the serious concerns raised regarding the amount and the impacts of agricultural air emissions, policies must be pursued and regulations must be enacted in order to make real progress in reducing these emissions and their associated environmental impacts.

I. Introduction

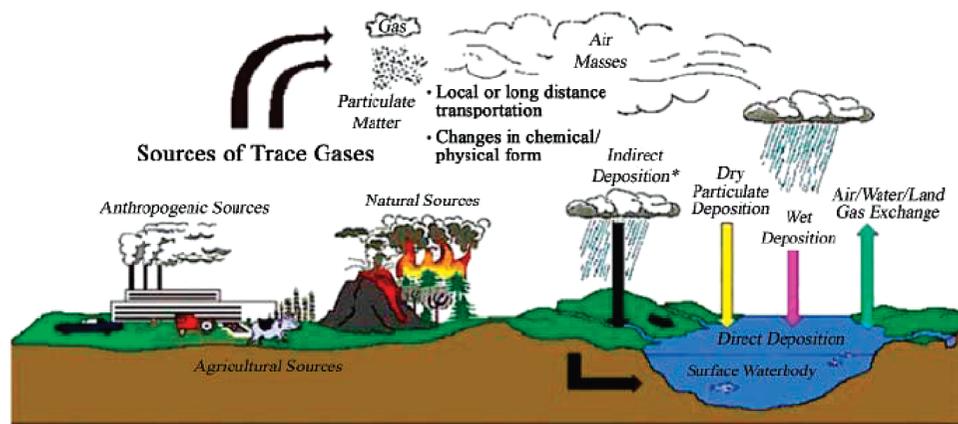
The world's population has grown from ~1.5 billion at the beginning of the 20th century to ~6.8 billion today. This population increase has been accompanied by the advent and growth of "intensive" agriculture, with associated impacts on the environment (1). During the next 50 years, the Earth's human population is predicted to increase to more than 9 billion, creating higher demand for agricultural commodities, both crop and animal. Without scientific research to inform policy decisions, there will likely be a parallel increase in environmental impacts associated with this future growth in agriculture (1–6).

Agronomists throughout the U.S. and Europe have sought to increase food production by increasing productivity. Farmers increased agricultural output significantly between the 1940s and the 1990s, capitalizing on increased availability of nitrogen fertilizer (the global production of fertilizer currently is more than 90 Tg of N yr⁻¹, compared to ~1 Tg only 50 years ago) (7, 8). Increased agricultural output is also the result of mechanization combined with the abandonment of traditional practices, better pesticides, cultivation of marginal land, availability of hybrid and genetically modified crop varieties, and improvements in production efficiency. Many of these innovations have been supported by public investment. Furthermore, inexpensive fossil fuels have been available for fertilizer production, for replacement of human labor by increased mechanization, and for transport of raw material and products.

In both the U.S. and Western Europe, the governmental agricultural policies encouraged intensification and commercial factors magnified this effect. Farmers increased agricultural intensity by the sustained use of chemical inputs, increasing field size, and higher animal stocking densities (i.e., concentrated animal feeding operations, CAFOs). Farmers discontinued traditional fallowing practices and crop rotations, resulting in a displacement of leguminous fodder crops with increased use of silage and maize for livestock. Specialization and intensification have resulted in a decrease in the number of farm holdings and the number of people employed in farming. This has been accompanied by a concentration of production, leading to less diversity of local agricultural habitats.

Growing public and regulatory concerns have recognized the emissions and discharges from agriculture and adverse impacts of agriculture on the quality of the air and water, and on soil, biodiversity, and the long-term sustainability of agricultural ecosystems (6). Public concerns about current and predicted impacts to the environment pressure farmers to reduce intensive agriculture. To develop policies to reduce environmental impacts from agriculture, we must understand the behavior of agricultural emissions and the subsequent transformations, transport, and fate of pollutants in the environment (Figure 1). Recognizing the growing needs in this research area, a number of governmental agencies, universities, and research organizations cosponsored an international workshop on agricultural air quality (9) during June 2006 (<http://www.esa.org/AirWorkshop>), to synthesize and assess existing measurements and modeling results and

* Corresponding author phone: (919) 515-7808; fax: (919) 515-7802; e-mail: Viney_Aneja@ncsu.edu.



*Indirect deposition is direct deposition to land followed by runoff or seepage through groundwater to a surface waterbody.

FIGURE 1. Atmospheric emissions, transport, transformation, and deposition of trace gases. Source: Aneja et al., 2006 (5).

to identify emerging research questions concerning agricultural air quality (4, 10).

This paper examines the state of the science for agricultural air quality, as well as future research opportunities for studying agriculture-related pollutants and their impacts on air quality, human health, and regional climate. We focus on ammonia and on the shortcomings of current air quality models applied to agriculture.

II. Agriculture and Its Contribution to Different Environmental Issues

U.S. agriculture is diverse, ranging from large, highly intensive, and specialized commercial holdings to subsistence (i.e., family owned) farming, using mainly traditional practices. Consequently, impacts on the environment vary in scale and intensity and may be positive or negative (1). However, increasing evidence shows that the greater size and intensity of farms and concentrated animal-feeding operations (CAFOs) increase the emissions of odorous compounds (e.g., organic acids) and trace gases (e.g., carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), nitrogen oxides (NO_x), ammonia (NH₃), and reduced sulfur compounds, such as hydrogen sulfide (H₂S)) to the atmosphere (4–6, 11, 13). For example, globally the livestock sector (beef and dairy cattle, swine, and poultry) is estimated to be responsible for ~18% of all greenhouse gas emissions measured in CO₂ equivalents, ~65% of anthropogenic nitrous oxide, ~37% of anthropogenic methane, and ~64% of anthropogenic ammonia (1). Globally, the livestock sector is a major driver of deforestation, as well as one of the leading drivers of land degradation, pollution, climate change, coastal sedimentation, and invasion of alien species (1, 6). In addition to these global environmental impacts, uncontrolled agricultural emissions in the United States will impact the ability of states to meet their legal obligations under the Clean Air Act. For example, NH₃ plays a significant role in PM_{2.5} formation, and increasing ammonia may enhance PM_{2.5} (aerosols with aerodynamic diameters of less than or equal to 2.5 μm) concentrations despite recent progress to lower emissions of sulfur oxides (SO_x) and NO_x. Ammonia-derived PM_{2.5} may challenge the stringent 24-h average National Ambient Air Quality Standard (NAAQS) for PM_{2.5} of 35 μg m⁻³ promulgated by the U.S. Environmental Protection Agency (4).

In the United States, air quality research in the past half-century has focused largely on NO_x, sulfur dioxide (SO₂), ozone (O₃), and particulate matter (PM_{2.5} and PM₁₀ (aerosols with aerodynamic diameters of less than or equal to 10 μm)). Limited attention has been given to reduced nitrogen-, sulfur-, and carbon-containing compounds. Compounds such as NH₃, hydrogen sulfide (H₂S), and volatile organic compounds (VOCs) play important roles in the formation of

criteria pollutants such as tropospheric O₃, SO₂, and PM_{2.5}, as well as in the acidification and eutrophication of ecosystems. These compounds interact in atmospheric reactions (e.g., gas-to-particle conversion, 14–18), are transported by winds, and return to the surface by wet and dry deposition (5, 10, 19). Many of these compounds have adverse effects on human health and the environment. Agriculture provides major sources of reduced gases and particulate matter during livestock production, fertilizer application, land use changes, and biomass burning (3–5). Approximately 90% of the global emission of NH₃ results from animal and crop agriculture (20), much of it from the U.S. and European countries (21–25).

There are no nationwide monitoring networks in the U.S. to quantify agricultural emissions of greenhouse gases (GHGs, (e.g., N₂O, CH₄, etc.)), NO_x, reduced sulfur compounds, VOCs, or NH₃. In contrast there is a large network in place to assess the changes in atmospheric chemistry associated with fossil fuel combustion. For instance, the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) has been monitoring the wet deposition of sulfate (SO₄²⁻), nitrate (NO₃⁻), and ammonium (NH₄⁺) since 1978 and currently has some 250 sites across the U.S. (<http://nadp.sws.uiuc.edu/>). Similarly, since 1987 the Clean Air Status and Trends Network (CASTNET) has been monitoring dry deposition of NO₃⁻, NH₄⁺, and HNO₃ (but not NH₃, NO) at 70 sites primarily in the eastern U.S. (<http://www.epa.gov/castnet/>).

Animal production results in emissions of hundreds of identified VOCs (26–31). These compounds are diverse, and include many acids, alcohols, aldehydes, amides, amines, aromatics, esters, ethers, halogenated hydrocarbons, hydrocarbons, ketones, nitriles, other nitrogen-containing compounds, phenols, sulfur-containing compounds, and steroids. Some of these compounds are responsible for unpleasant odors and for impacts on the comfort, health, and production efficiency of animals and humans (32, 33).

Hydrogen sulfide (H₂S), a major emission from animal agriculture, is a colorless, potentially lethal gas released from swine manure decomposition (34). It is produced as manure decomposes anaerobically, resulting from the mineralization of organic sulfur compounds as well as the reduction of oxidized inorganic sulfur compounds such as sulfate by sulfur-reducing bacteria (35). The U.S. Center for Disease Control (CDC) warns that brief exposures to high concentrations (>500 ppm) can cause unconsciousness or death (36). Campagna et al. (37) have reported a correlation between elevated ambient H₂S concentrations and hospital visits for respiratory diseases. Donham et al. (38) reported that hydrogen sulfide and “manure gas” appeared to be the main toxic substance associated with death and illness for people

TABLE 1. U.S. and Europe Air Pollutant Emission Estimates (million tons/yr) (1 ton = 2000 pounds)

	CO	NO _x	PM _{2.5}	PM ₁₀	SO ₂	VOC	Pb	NH ₃
1970	197.3	26.9	2.3 (1990)	12.2	31.2	33.7	0.221	1.9
2005	89	19	2	2	15	16	0.003	2.6
percent change (U.S.)	-55%	-29%	-13%	-84%	-52%	-53%	-99%	+27%
percent change (Europe)2005-1990	-50%	-31%	-53%	-45%	-66%	-41%	-87%	-20%

^a Source: <http://www.epa.gov/airtrends/econ-emissions.html> (42, 47). European data: European Environment Agency and EMEP (<http://www.eea.europa.eu>).

with acute exposure to gases emanating from liquid manure. With an odor threshold ranging from 0.0005 to 0.3 ppm (36), it is one of the primary gases released from swine facilities causing odor complaints due to its characteristic "rotten egg" smell. H₂S is the major sulfur compound emitted from concentrated animal-feeding operations (12, 13, 39), but we know little about the emission of other gaseous sulfur compounds, such as methyl mercaptan (CH₃SH), dimethyl sulfide (DMS (CH₃)₂S), dimethyl disulfide (DMDS (CH₃)₂S₂), carbonyl sulfide (COS), and carbon disulfide (CS₂).

Nitrous oxide is a greenhouse gas with an atmospheric lifetime of approximately 120 years. Nitrous oxide is about 310 times more effective in trapping heat in the atmosphere than CO₂ over a 100-year period (40). It is produced naturally in soils through the microbial processes of denitrification and nitrification. These natural emissions of N₂O can be increased by a variety of agricultural practices and activities, including the use of synthetic and organic fertilizers, production of nitrogen-fixing crops, cultivation of organic soils, and the application of livestock manure to croplands and pasture. Nitrous oxide emissions from croplands fertilized for the production of biofuels can negate all of the benefits of this renewable source of energy on the Earth's climate (41). Agricultural sources (both crop and animal production) account for ~72% of N₂O emissions in the U.S. (42).

Inadvertent additions of nitrogen to soils can also result in N₂O emissions. Indirect emissions occur when applied fertilizer or manure nitrogen volatilizes as ammonia and oxides of nitrogen, which are then deposited in downwind regions in the form of particulate ammonium, nitric acid, and oxides of nitrogen. Surface runoff and leaching of applied nitrogen into groundwater and surface waters can also result in indirect N₂O emissions from downstream ecosystems (43).

Globally, agriculture (animal and crop) is the most important source of anthropogenic methane. Among domesticated livestock, ruminant animals (cattle, buffalo, sheep, goats, and camels) produce significant amounts of methane as part of their normal digestive process (42). The anaerobic decomposition of organic material in livestock manure also releases methane, especially when manure is stored in liquid form, in lagoons or holding tanks. Lagoon systems are typical for most large-scale hog operations in the U.S. (42). Anthropogenic methane emissions from livestock account for ~37% of total global emissions. Rice paddies are the primary source of methane in crop agriculture (44).

Primary emissions of particles from agriculture in the U.S. contribute about 16% to the PM_{2.5} emissions, and ~18% to PM₁₀ emissions. However, there is no estimate for the secondary formation of PM_{fine} from precursor gases emitted from agriculture. Current investigations show that PM emissions from agriculture in regions of intensive ammonia emission may have been previously underestimated, and a large part of the gap between modeled and measured PM concentrations might be explained by previously underestimated agricultural sources (45).

Ambient PM_{2.5} results from direct particle emissions (e.g., soil dust) and secondary particles (generated by atmospheric reactions of precursor gas emissions). The major precursor gases include SO₂, NO_x, VOCs, and NH₃. The mass of ambient PM_{2.5} is thus a mixture composed mostly of sulfate (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), organic carbon (OC), black carbon (BC), and soil dust. A considerable and growing body of evidence shows an association between adverse health effects and exposure to ambient levels of PM (46). The primary PM_{2.5} emissions from agricultural sources in the U.S. are approximately 946 thousand tons/year (1 ton = 2000 lbs) (47), composed of emissions from fertilizer and livestock of approximately 4 thousand tons, agricultural emissions from tilling and harvesting of approximately 717 thousand tons, and agricultural emissions from fires of approximately 225 thousand tons. The total PM_{2.5} emissions from all sources in the U.S. is approximately 6,031 thousand tons/year.

The reactions between NH₃, sulfuric acid (H₂SO₄), nitric acid (HNO₃), hydrochloric acid (HCl), and water (H₂O) are the most important equilibrium reactions for gas/particle partitioning and the formation of ammonium (NH₄⁺) salts, which make up ~20% of the PM_{2.5} in the atmosphere (4, 5, 14-17, 48, 49). Once formed, these particles act as cloud condensation nuclei, which affect the Earth's radiation budget and its climate through cloud formation, lifetime, and precipitation. The aqueous phase chemistry of NH₃ may also provide a mechanism for reduced nitrogen to repartition from larger particles to small particles, thus forming new particles in ultrafine mode (<0.1 μm aerodynamic diameter) (50).

The PM₁₀ emissions for agriculture (47) are approximately 4,032 thousand tons/year. These agriculture emissions consist of PM₁₀ from crop tilling and livestock dust emissions of approximately 3,751 thousand tons, 265 thousand tons for agricultural field burning, and 15 thousand tons from livestock waste and fertilizer application. This compares to total U.S. PM₁₀ primary emissions of approximately 21,919 thousand tons in 2002.

III. NH₃ Emission Control and Policy Implications

In the U.S., there are no federal regulations that control ammonia emissions from agricultural operations. States have generally refrained from regulating emissions from any agricultural sources, even though such regulation may be permitted (51). The extensive Clean Air Act permitting system and pollution control measures applied to SO₂, NO_x, and anthropogenic VOCs have not been extended to ammonia by EPA (Table 1). Currently, incentives to reduce criteria pollutants from agriculture aim primarily at preventing soil loss by wind erosion processes rather than at reducing gaseous emissions (52, 53).

In contrast, in Europe, health and environmental concerns about agriculturally emitted air pollutants have led regulators and policy makers to implement mitigation strategies for ammonia (Table 1). For example, in The Netherlands, livestock production must meet stringent targets for NH₃

emission (54, 55). However, in the U.S., both the USDA and U.S. EPA have shown a preference for voluntary mitigation strategies for ammonia (i.e., Best Management Practices, BMPs), some of which are beginning to be implemented (4). Nevertheless, there are no national ambient air quality standards (NAAQS) for ammonia and hydrogen sulfide in the U.S., and applicable regulatory provisions for emissions from CAFOs are weak. Separately, some states (e.g., California) are developing regulations to curb emissions of ammonia and hydrogen sulfide.

The reporting requirements within the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA, also known as the Superfund program) and the Emergency Planning and Community Right-to-Know Act (EPCRA) for releases of NH_3 and H_2S now have an exemption applicable to emissions from CAFOs. The application of CERCLA and EPCRA reporting requirements to CAFOs has been controversial and over the past few years there have been legislative attempts to exempt manure management from these regulations. Although these legislative efforts have repeatedly failed, U.S. EPA recently finalized a limited administrative reporting exemption for releases of hazardous substances from animal waste at farms (56). The exemption became effective on January 20, 2009 and exempts all farms that have air releases from animal waste from CERCLA Section 103. The final rule also exempts farms that release reportable quantities of hazardous substances from reporting under EPCRA Section 304 if they confine fewer animals than a large CAFO as defined in the NPDES regulations. In addition, some states are implementing mitigation measures. For example, both Minnesota and Texas have ambient air quality standards for H_2S , and in 1999 North Carolina was one of the first states in the U.S. to adopt rules for odor control from swine farms. U.S. policymakers should follow the lead of their counterparts in western Europe by introducing regulations in the U.S.

Although little attention has been given to reducing NH_3 emissions in the U.S., this has been an important policy issue in Europe. A number of studies have been performed to estimate the efficiency of various abatement options (24, 55, 57). Multidisciplinary modeling approaches combine information about environmental impacts, biophysical processes, and agricultural operations (e.g., soil, land use, crop, fertilizer, irrigation). For example, Cowell and Apsimon (58) have developed the Model for the Assessment of Regional Ammonia Cost Curves for Abatement Strategies (MARACCAS) to assess the cost-effectiveness of potential abatement measures and to design efficient abatement strategies. McCubbin et al. (57), employing the S-R (Source-Receptor) matrix AQM (air quality model), suggested that reducing livestock NH_3 emissions by 10% could lead to particulate-related health benefits of over \$4 billion yr^{-1} in the EU. The Regional Air Pollution Information and Simulation (RAINS) model includes several options to control NH_3 , including lowering the nitrogen content in feed, air purification, improvements in animal housing, covered storage of manure, low NH_3 application of manure, urea substitution, and stripping and absorption techniques in fertilizer industry (59). Abatement of NH_3 may also adversely impact (i.e., increase) the emissions of CH_4 and N_2O (60).

Diffusive sources have been studied by the plume method (61), which can be used to estimate emission factors (a representative value that relates the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant). In past years, wet-denuder techniques have been used to determine the cross-wind integrated concentration (62). Currently tunable diode lasers and quantum cascade lasers are used for NH_3 , N_2O , and CH_4 emission measurements (63). Methane plume measurements carried out within the Greengrass EU project (<http://www.clermont.inra.fr/greengrass/>) showed that the emission

factor for dairy cows in The Netherlands was higher than that in the national methane inventory. Evaluation of this emission factor with more accurate data on animal weight and milk production shows that emissions are 20% higher than the traditional factor.

More recently The GAINS (greenhouse gas - air pollution interactions and synergies) model (64), which is an integrated assessment model, has been used to allocate emissions across economic sectors. GAINS brings together information on the sources and impacts of air pollutants and greenhouse gases and their interactions. GAINS also includes data on economic development, the structure, potential control, and costs of emission sources, the formation and dispersion of pollutants in the atmosphere, and an assessment of the environmental impacts of pollution. GAINS addresses air pollution impacts on human health from fine particulate matter and ground-level ozone, vegetation damage caused by ground-level ozone, the acidification of terrestrial and aquatic ecosystems, and the effects of excess nitrogen deposition on soils, in addition to the mitigation of greenhouse gas emissions. GAINS describes the interrelations among these multiple effects and the range of pollutants (SO_2 , NO_x , PM, NMVOC, NH_3 , CO_2 , CH_4 , N_2O , F-gases) that contribute to air quality in Europe.

There are large uncertainties in current agricultural air quality modeling as a result of a number of factors including (1) inaccurate emission inventories and activity data (e.g. when, where, and what kind of manure/fertilizer is applied); (2) inaccurate meteorological data (e.g., temperature, wind speed, wind direction, and precipitation); (3) a lack of detailed information on terrain characteristics and land use at a fine scale (e.g., topography, surface roughness, and vegetation); (4) missing or inadequate model treatments of chemical and physical processes (e.g., gas- and aqueous-phase chemistry for NH_3 and hydrogen sulfide, gas/particle partitioning, aerosol dynamics, and dry and wet deposition); (5) inability to simulate both the short-range dispersion and deposition of NH_3 near the ground and the long-range transport and fate of NH_4^+ at higher elevations downwind of sources; (6) high uncertainty in the dry deposition of reactive nitrogen, sulfur, and carbon compounds emitted from agriculture; and (7) a paucity of observations of emissions, concentrations, and deposition suitable for model verification and evaluation. Reconciling modeled results with measurements is further complicated by the weather, which has a profound effect on ambient concentrations and dry deposition. Small changes in temperature, wind speed, or humidity may change the ambient concentration and dry deposition regardless of emissions. Given predictions of global climate change, the exponential increase in trace gas emissions with temperature due to gas/solution partitioning is particularly important.

The most advanced technologies for reducing ammonia (e.g., manure injection in soil systems, low emission housing systems, etc.) are found in The Netherlands, Denmark, and the UK (65). The Dutch mineral bookkeeping system at the farm level keeps track of all the nitrogen flows, including nitrogen deposition, and provides a helpful tool to decrease farm-level nitrogen surplus (input minus output). Since the introduction of the system of mineral bookkeeping in The Netherlands in 1998, there has been a significant reduction of the nitrogen surplus in the agricultural sector due to a reduction of the use of inorganic fertilizers (55). Success in reducing ammonia emissions has also been achieved through requirements to reduce the volatilization from manure and urea and indirectly as a result of a quota on milk production, a reduction in feed nitrogen, and improved management of nitrogen on the farm. There are two major options to reduce nitrogen and ammonia emissions: (1) reducing the inputs and at the same time increasing effective use, and (2) reducing emissions through technology (55). Management options are

farm-specific. Technology provides more general options, which must be assessed for potential pollutant swapping, i.e., the increased emission of one pollutant resulting from abating another. An example of pollutant swapping is the increased nitrate leaching as the result of manure injection without reducing the nitrogen application rate (66). Manure injection systems reduce the contact surface of manure slurry with the atmosphere after application to decrease emissions and encompass systems of direct slurry injection into the soil, digging of small ditches filled with manure, or direct under-plowing of manure after application.

Currently the models predict changes in concentrations reasonably well (67), but many models predict 25–30% lower concentrations than measured (66, 68). The gap may result from overestimates of dry deposition and the underestimation of emissions during land application of manure by injection.

Promising results have been reported in the U.S. for reducing ammonia from swine manure through the use of “engineered systems”, i.e., a treatment plant with solid–liquid separation (69). Szogi (70) reported a 73% reduction in ammonia emissions from the implementation of such a system. Vanotti (71) found that when manure was processed in such a system, greenhouse gas emissions were reduced by 98.8%, and additional income of \$9,100 to \$27,500/year (approximately \$0.91/finished pig) was generated from the sale of byproducts. In addition, for row crop production, when organic fertilizers are applied with gypsum, they can reduce ammonia volatilization by ~11% (72).

“Environmentally superior technology” (EST) represents a recent initiative in North Carolina to develop alternatives to lagoon treatment and land application of swine manure. EST focuses on impacts of animal waste to surface and groundwater, by emissions of ammonia, odor, and disease-transmitting pathogens, and heavy metal contamination of soil and groundwater. Five technologies have been shown to reduce these impacts: a solids separation/nitrification-denitrification/soluble phosphorus removal system; a thermophilic anaerobic digester system; a centralized composting system; a gasification system; and a fluidized bed combustion system. Economic data compiled for all EST systems showed annualized (10-year) costs of retrofitting existing swine farms ranged between \$90 and over \$400 per 1000 lbs. steady-state live weight (53). These ESTs are now being modified to handle manure from other animal agricultural systems (e.g., dairy and poultry waste).

IV. Current Topics of Research for Agriculture

Research should now focus on quantifying agricultural point and nonpoint sources of air pollutants; the biosphere–atmosphere exchange of reactive nitrogen (e.g., NH_3 , NO_x , N_2O , etc.), sulfur (e.g., H_2S), and VOCs; the quantification of the primary and secondary emissions of PM; the gas-to-particle conversions; the constituents and dynamics of odor (66); and greenhouse gas emissions (33, 41, 44).

We have only limited understanding of the biosphere–atmosphere exchange of agriculturally emitted trace gases. After deposition, NH_3 can be re-emitted in areas of lower ambient concentration, which shifts the equilibrium toward gaseous forms. The atmosphere–biosphere exchange is largely driven by this equilibrium, which shows high variation in space and time (73–75). The challenge will be to model fluxes of gases and particulate matter on the regional or landscape scale, where both emission and deposition take place at the same time (76).

The contribution of agriculture to PM concentrations, both primary PM_{10} emissions as well as secondary formation of $\text{PM}_{2.5}$ (i.e., PM_{fine}) with NH_3 as precursor, remains a challenge (5). Erisman and Schaap (2003) concluded that $\text{PM}_{2.5}$

concentrations can best be reduced when emissions of SO_2 , NO_x , and NH_3 are all reduced simultaneously. $\text{PM}_{2.5}$ reduction strategies focused on SO_2 and NO_x while ignoring NH_3 are not as effective. However, NH_3 reductions alone were somewhat effective in reducing $\text{PM}_{2.5}$. Similar conclusions were reported in a modeling study by Meng et al. (77) in the U.S.

Agriculture is an important sector contributing to environmental effects and air quality. Agricultural air pollutants contribute to human health problems through exposure to ammonia, hydrogen sulfide, toxic organic compounds, pesticides, and particulate matter. Agricultural air pollution contributes to climate change in the form of greenhouse gas emissions and aerosols. Agricultural air pollution also contributes to odor. After deposition of reactive nitrogen, eutrophication and acidification can result and biodiversity is endangered.

Ammonia, in particular, plays a role in a host of environmental problems (e.g., air quality, odor, climate change, soil acidification, eutrophication, biodiversity), often through interactions with other compounds in the atmosphere. The central challenge is how to optimize the use of nitrogen to sustain human life while minimizing its negative impacts on the environment and human health.

Production agriculture has adopted modern technologies and science to maximize productivity, but it has not yet been subjected to the same environmental regulations that other modern industries must obey. Regulations and policies should also require that CAFOs and crop production systems use all of the practical methods to reduce ammonia and other air emissions. The potential health and environmental risks of intensified modern agriculture demand that we develop emission abatement policies based on best available science (6, 10, 69).

Reducing uncertainties presents significant research challenges and charts the direction of research for the next decade or beyond. Resolving them will have important policy implications on local to global scales and will profoundly improve air quality, human health, the agricultural environments, and biodiversity. Progress on these challenges will require an integrated and multidisciplinary effort both nationally and globally from scientists, engineers, policy-makers, managers, and the public.

Acknowledgments

We acknowledge support from the Cooperative State Research, Education, and Extension Service (CSREES), USDA National Research Initiative Competitive Grants Program, Contracts 2004-35112-14253 and 2005-35112-15377, the National Science Foundation (Grant ATM-0528420), and the EU Integrated project NitroEurope, the COST Action 729 and the European Science Foundation Program NinE. We also thank North Carolina State University; the Center for the Environment, Purdue University; the Nicholas School of the Environment and Earth Sciences, Duke University; the Ecological Society of America; Environmental Defense Fund; the Indo-US Science and Technology Forum; the Kenan Institute for Engineering, Technology and Science; the North Carolina Department of Environment and Natural Resources, Division of Air Quality; the North Carolina Water Resources Research Institute; the Phosphate and Potash Institute, the Fertilizer Institute; the United States Department of Agriculture, the Agricultural Research Service; and the United States Environmental Protection Agency for their support. We also thank Professor Ralph Cicerone, President, National Academy of Sciences, for the workshop keynote address. We thank Joe Rudek, Environmental Defense; Sally Shaver, U.S. Environmental Protection Agency; James P. Longest, Jr. “Ryke”, NC Special Deputy Attorney General; Michele Merkel, Waterkeeper Alliance; Avinash Kar and Luke Cole, Center on

Race, Poverty & the Environment; and Zbigniew Klimont, International Institute for Applied Systems Analysis (IIASA), Austria for review of the manuscript. We greatly appreciate the time and interest of the many researchers, scientists, engineers, policy makers, interest groups, and graduate students who shared their knowledge and expertise to make the *Workshop on Agricultural Air Quality: State of the Science* (<http://www.esa.org/AirWorkshop>) a success. Finally, the thoughtful comments and suggestions by the reviewers are greatly appreciated.

Literature Cited

- FAO. *Livestock's Long Shadow*; United Nations, Food and Agriculture Organization, 2006; http://www.virtualcentre.org/en/library/key_pub/longshad/A0701E00.pdf.
- Smil, V. *Carbon-Nitrogen-Sulfur: Human Interference in Grand Biospheric Cycles*; Plenum Press: New York, 459, 1985.
- Millenium Ecosystem Assessment*; United Nations, Food and Agriculture Organization, 2005; <http://www.millenniumassessment.org/en/Reports.aspx>.
- Aneja, V. P., Schlesinger, W., Knighton, R., Jennings, G., Niyogi, D., Gilliam, W., Duke, C., Eds. *Workshop on Agricultural Air Quality: State of the Science*; 27695-7603, ISBN 0-9669770-4-1; North Carolina State University: Raleigh, NC, 2006; 1314 pp.
- Aneja, V. P.; Schlesinger, W. H.; Niyogi, D.; Jennings, G.; Gilliam, W.; Knighton, R. E.; Duke, C. S.; Blunden, J.; Krishnan, S. Emerging national research needs for agricultural air quality. *Eos. Trans., Am. Geophys. Union* **2006**, *87* (3), 25–29.
- Aneja, V. P.; Schlesinger, W.; Erisman, J.W. Farming pollution. *Nat. Geosci.* **2008**, *1*, 409–411.
- The Fertilizer Institute, 2000; <http://tfi.org/worconch.pdf>.
- International Fertilizer Industry Association (IFA). *Summary Report: World Agriculture and Fertilizer Demand, Global Fertilizer Supply and Trade 2004–2005*; IFA: Paris, France, 2004.
- WAAQ. Workshop on Agricultural Air Quality. Sponsored by U.S. Department of Agriculture - USDA, the National Science Foundation - NSF, the U.S. Environmental Protection Agency - EPA, the North Carolina Division of Air Quality - NCDQA, North Carolina State University - NCSU, Duke University, Purdue University, the Air and Waste management Association - A&WMA, and the Ecological Society of America, 2006; <http://www.esa.org/AirWorkshop>.
- Aneja, V. P.; Blunden, J.; Roelle, P. A.; Schlesinger, W. H.; Knighton, R.; Niyogi, D.; Gilliam, W.; Jennings, G.; Duke, C. S. Workshop on Agricultural Air Quality: State of the Science. *Atmos. Environ.* **2008**, *42*, No. 14, 3195–3208.
- National Research Council, NRC. *Air Emission from Animal Feeding Operations: Current Knowledge, Future Needs*; The National Academies Press: Washington, DC, 2003; 263 pp.
- Blunden, J.; Aneja, V. P. Characterizing ammonia and hydrogen sulfide emissions from a swine waste treatment lagoon in North Carolina. *Atmos. Environ.* **2008**, *42*, 3277–3290.
- Blunden, J.; Aneja, V. P.; Westerman, P. W. Measurement and analysis of ammonia and hydrogen sulfide emissions from a mechanically ventilated swine confinement building in North Carolina. *Atmos. Environ.* **2008**, *42* (14), 3315–3331.
- Ansari, A. S.; Pandis, S. N. Response of inorganic PM to precursor concentrations. *Environ. Sci. Technol.* **1998**, *32*, 2706–2714.
- Adams, P. J.; Seinfeld, J. H.; Koch, D. M. Global concentrations of tropospheric sulphate, nitrate and ammonium aerosol simulated in a general circulation model. *J. Geophys. Res.* **1999**, *104* (13), 791813, 823.
- Baek, B. H.; Koziel, J.; Aneja, V. P. A preliminary review of Gas-to-Particle Conversion, monitoring, and modeling efforts in the USA. *Int. J. Global Environ. Iss.* **2006**, *6* (2/3), 204–230.
- Baek, B. H.; Aneja, V. P. Measurement and analysis of the relationship between ammonia, acid gases, and fine particles in Eastern North Carolina. *J. Air Waste Manage. Assoc.* **2004**, *54*, 623–633.
- Baek, B. H.; Aneja, V. P.; Tong, Q. Chemical coupling between ammonia, acid gases, and fine particles. *Environ. Pollut.* **2004**, *129*, 89–98.
- Hicks, B. B.; Draxler, R. R.; Albritton, D. L.; Fehsenfeld, F. C.; Hales, J. M.; Meyers, T. P.; Vong, R. L.; Dodge, M.; Schwartz, S. E.; Tanner, R. L.; Davidson, C. I.; Lindberg, S. E.; Wesely, M. L. *Atmospheric Processes Research and Processes Model Development*; State of Science/Technology, Report No. 2; National Acid Precipitation Assessment Program, 1989.
- Schlesinger, W. H.; Hartley, A. A global budget for atmospheric NH₃. *Biogeochemistry* **1992**, *15*, 191–211.
- Dentener, F. J.; Crutzen, P. J. A three-dimensional model of global ammonia cycle. *J. Atmos. Chem.* **1994**, *19*, 331–369.
- Bouwman, A. F.; Lee, D. S.; Asman, W. A. H.; Dentener, F. J.; VanderHoek, K. W.; Olivier, J. G. J. A global high-resolution emission inventory for ammonia. *Global Biogeochem. Cycles* **1997**, *11*, 561–587.
- Bouwman, A. F.; Boumans, L. J. M.; Bates, N. H. Estimation of global NH₃ volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Global Biogeochem. Cycles* **2002**, *16*, doi: 10.1029/2000GB001389.
- Van der Hoek, K. W. Estimating ammonia emission factors in Europe: summary of the work of the UNECE ammonia expert panel. *Atmos. Environ.* **1998**, *32*, 315–316.
- Hutchings, N. J.; Sommer, S. G.; Andersen, J. M.; Asman, W. A. H. A detailed ammonia emission inventory for Denmark. *Atmos. Environ.* **2001**, *35*, 1959–1968.
- Tamminga S. Gaseous pollutants by farm animal enterprises In *Farm Animals and the Environment*; Phillips, C., Piggins, D., Eds.; CAB International: Wallingford, U.K, 1992; pp 345–357.
- Hartung, J.; Phillips, V. R. Control of gaseous emissions from livestock buildings and manure stores. *J. Agric. Eng. Res.* **1994**, *57*, 173–189.
- Zahn, J. A.; Hatfield, J. L.; Do, Y. S.; DiSpirito, A. A.; Laird, D. A.; Pfeiffer, R. L. Characterization of volatile organic emissions and wastes from a swine production facility. *J. Environ. Qual.* **1997**, *26*, 1687–1696.
- Zahn, J. A.; DiSpirito, A. A.; Do, Y. S.; Brooks, B. E.; Cooper, E. E.; Hatfield, J. L. Correlation of human olfactory responses to airborne concentrations of malodorous volatile organic compounds emitted from swine effluent. *J. Environ. Qual.* **2001**, *30*, 624–634.
- Schiffman, S. S.; Bennett, J. L.; Raymer, J. H. Quantification of odors and odorants from swine operations in North Carolina. *Agric. Forest Meteorol.* **2001**, *10*, 213–240.
- Blunden, J.; Aneja, V. P.; Lonneman, W. A. Characterization of non-methane volatile organic compounds at swine facilities in eastern North Carolina. *Atmos. Environ.* **2005**, *39*, 6707–6718.
- Schiffman, S. S.; Walker, J. M.; Dalton, P.; Lorig, T. S.; Raymer, J. H.; Shusterman, D.; Williams, C. M. Potential health effects of odor from animal operations, wastewater treatment, and recycling of byproducts. *J. Agromed.* **2000**, *7* (1), 7–81.
- Concentrated animal feeding operations air quality study*; Iowa, 2002; www.public-health.uiowa.edu/ehsr.
- U.S. Environmental Protection Agency. *Toxicological Review of Hydrogen Sulfide (CAS No. 7783-06-4)*; EPA/635/R-03/005; U.S. EPA: Washington, DC, 2003.
- U.S. Environmental Protection Agency. Chapter 13, Non-Water Quality Impact Estimates for Animal Feeding Operations. In *Proposed Rule Development Document for Concentrated Animal Feeding Operations (CAFOs)*; EPA-821-R-01-003; U.S. EPA: Washington, DC, 2001; http://www.epa.gov/npdes/pubs/cafo_nonwaterquality.pdf.
- Agency for Toxic Substances and Disease Registry (ATSDR). *Toxicological profile for hydrogen sulfide (Draft for Public Comment)*; U.S. Department of Health and Human Services, Public Health Service: Atlanta, GA, 2004.
- Campagna, D.; Kathman, S. J.; Pierson, R.; Inserra, S. G.; Phipper, B. L.; Middleton, D. C.; Zarus, G. M.; White, M. C. Ambient hydrogen sulfide, total reduced sulfur, and hospital visits for respiratory diseases in northeast Nebraska, 1998–2000. *J. Expos. Anal. Environ. Epidemiol.* **2004**, *14*, 180–187.
- Donham, K. J.; Knapp, L. W.; Monson, R.; Gustafson, K. Acute toxic exposure to gases from liquid manure. *J. Occup. Med.* **1982**, *24* (2), 142–145.
- Blunden, J.; Aneja, V. P.; Overton, J. H. Modeling hydrogen sulfide emissions across the gas-liquid interface of an anaerobic swine waste treatment storage system. *Atmos. Environ.* **2008**, *42* (22), 5602–5611.
- Intergovernmental Panel on Climate Change (IPCC). Fourth Assessment Report, 2007; <http://www.ipcc.ch/ipccreports/ar4-syr.htm>.
- Crutzen, P. J.; Mosier, A. R.; Smith, K. A.; Winiwarter, W. N₂O release from agro-biofuel production negates global warming reduction by replacing fossil fuels. *Atmos. Chem. Phys. Discuss.* **2007**, *7*, 11191–11205.
- U.S. Environmental Protection Agency. *2005 US Emissions Inventory 2005: Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2003*; U.S. EPA: Washington, DC; <http://www.epa.gov/nitrous1/sources.html>.
- Schlesinger, W. H. On the fate of anthropogenic nitrogen. *Proc. Natl. Acad. Sci.* **2009**, *106*, 203–208.

- (44) Zhuang, Q.; Melack, J. M.; Zimov, S.; Wlaler, K. M.; Butenhoff, C. L.; Khalil, M. A. K. Global methane emissions from wetlands, rice paddies, and lakes. *EOS* **2009**, *90* (5), 37–38.
- (45) Wu, S. Y.; Krishnan, S.; Zhang, Y.; Aneja, V. P. Modeling atmospheric transport and fate of ammonia in southeast U.S., Part I: Evaluation of meteorological and chemical predicitions. *Atmos. Environ.* **2008**, *42*, 3419–3436.
- (46) McMurry, P.; Shepherd, M.; Vickery, J., *Particulate Matter Science for Policy Makers*; 510; Cambridge University Press, 2004.
- (47) U.S. Environmental Protection Agency. *2002 National Emissions Inventory Version 1*; EPA-454/R-05-001; U.S. EPA: Washington, DC.
- (48) Aneja, V. P.; Wang, B.; Tong, Q.; Kimball, H.; Steger, J. Characterization of major chemical components of fine particulate matter in North Carolina. *J. Air Waste Manage. Assoc.* **2006**, *56*, 1099–1107.
- (49) Hu, J. L.; Wu, S. Y.; Zhang, Y.; Aneja, V. P. Modeling atmospheric transport and fate of ammonia in Southeast U.S., Part II: Effect of ammonia emissions on fine particulate matter formation. *Atmos. Environ.* **2008**, *42*, 3437–3451.
- (50) Seinfeld, J. H.; Pandis, S. N. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*; John Wiley & Sons, Inc.: NJ, 2006; p 1203.
- (51) Ruhl, J. B. Farms, their environmental harms, and environmental law. *27 Ecology Law Quarterly* **2000**, *263*, 1–89.
- (52) General Assembly of North Carolina, Session 2007, Senate Bill 1465; <http://www.ncleg.net/Sessions/2007/Bills/Senate/HTML/S1465v7.html>.
- (53) Williams, C. M. Development of environmentally superior technologies in the US and policy. *Bioresour. Technol.* **2009**, doi: 10.1016/j.biortech.2009.01.067.
- (54) Lekkerkerk, L. J. A. Implications of DUTCH ammonia policy on the livestock sector. *Atmos. Environ.* **1998**, *32* (3), 581–587.
- (55) Erisman, J. W.; Domburg, N.; de Vries, W.; Kros, H.; de Haan, B.; Sanders, K. The Dutch N-cascade in the European perspective. *Sci. China* **2005**, *48*, 827–842.
- (56) U.S. Environmental Protection Agency. *CERCLA/EPCRA Administrative Reporting Exemption for Air Releases of Hazardous Substances From Animal Waste at Farms*; Federal Register, Vol. 73, No. 244, Thursday, December 18, 2008/ Rules and Regulations (40 CFR Parts 302 and 355) 2008; pp 76948–76960.
- (57) Mccubbin, D. R.; Apelberg, B. J.; Roe, S.; Divita, F. Livestock ammonia management and particulate-related health benefits. *Environ. Sci. Technol.* **2002**, *36*, 1141–1146.
- (58) Cowell, D. A.; Apsimon, H. M. Cost-effective strategies for the abatement of ammonia emissions from European agriculture. *Atmos. Environ.* **1998**, *32*, 573–580.
- (59) Klimont, Z. *Ammonia emissions, abatement technologies and related costs for Europe in the RAINS model*; Interim Report IR-01; International Institute for Applied Systems Analysis (IIASA): Laxenburg, Austria, 2001.
- (60) Brink, C.; Kroeze, C.; Klimont, Z. Ammonia abatement and its impact on emissions of nitrous oxide and methane -- Part 2: application for Europe. *Atmos. Environ.* **2001**, *35*, 6313–6325.
- (61) Hensen, A.; Scharff, H. Methane emission estimates from landfills obtained with dynamic plume measurements. *Water, Air Soil Pollut.* **2001**, (focus 1), 455–464.
- (62) Erisman, J. W.; Otjes, R.; Hensen, A.; Jongejan, P.; van den Bulk, P.; Khlystov, A.; Slanina, J. Instrument development and application in studies and monitoring of ambient ammonia. *Atmos. Environ.* **2001**, *35*, 1913–1922.
- (63) Hensen, A.; Groot, T. T.; Van den Bulk, W. C. M.; Vermeulen, A. T.; Olesen, J. E.; Schelde, K. Dairy farm CH₄ and N₂O emissions from one square meter to the full farm scale. *Agric. Ecosyst. Environ.* **2006**, *112*, 146–152.
- (64) Klimont, Z.; Brink, C. *Modelling of emissions of air pollutants and greenhouse gases from agricultural sources in Europe*; Interim Report IR-04-048; International Institute for Applied Systems Analysis (IIASA): Laxenburg, Austria, 2004.
- (65) Erisman, J. W.; Bleeker, A.; Hensen, A.; Vermeulen, A. Agricultural air quality in Europe and the future perspectives. *Atmos. Environ.* **2008**, *42*, 3209–3217.
- (66) Erisman, J. W.; Bleeker, A.; van Jaarsveld, H. Atmospheric deposition of ammonia to semi-natural vegetation in the Netherlands-methods for mapping and evaluation. *Atmos. Environ.* **1998**, *32*, 481–489.
- (67) Van Jaarsveld, J. A. *The Operational Priority Substances Model: Description and Validation of OPS-pro 4.1*; RIVM Report nr. 500045001/2004; National Institute of Public Health and Environment: The Netherlands, 2004.
- (68) Smits M. C. J.; van Jaarsveld, J. A.; Mokveld, L. J.; Vellinga, O.; Stolk, A.; Hoek, K. W.; van der Pul, W. A. J. 2005VELD-project: a detailed inventarisatie of ammonia emissions and concentrations in an agricultural area. ISBN: 9067549193. RIVM rapport 500033002; A&F Report 429; A&F: Wageningen, the Netherlands; 183 p in Dutch.
- (69) Aneja, V. P.; Arya, S. P.; Rumsey, I. C.; Kim, D.-S.; Bajwa, K. S.; Williams, C. M. Characterizing ammonia emissions from swine farms in eastern North Carolina: Reduction of emissions from water-holding structures at two candidate superior technologies for waste treatment. *Atmos. Environ.* **2008**, *42*, 3291–3300.
- (70) Szögi, A. Reduction of ammonia emissions from swine lagoons using alternative wastewater technologies. *The Workshop on Agricultural Air Quality: State of Science, 4 June 2006*; The Ecological Society of America, 2006; pp 1155–1160.
- (71) Vanotti, M. Greenhouse gas emission reduction and credits from implementation of aerobic manure treatment systems in swine farms *The Workshop on Agricultural Air Quality: State of Science, 4 June 2006*; The Ecological Society of America, 2006; pp 1178–1185.
- (72) Model, A. Ammonia and trace gas emissions from organic fertilizers amended with gypsum *The Workshop on Agricultural Air Quality: State of Science, 4 June 2006*; The Ecological Society of America, 2006; pp 923–929.
- (73) Sutton, M. A.; Burkhardt, J. K.; Guerin, D.; Nemitz, E.; Fowler, D. Development of Resistance models to describe measurements of bi-directional ammonia surface-atmosphere exchange. *Atmos. Environ.* **1998**, *32* (Special Issue), 473–480.
- (74) Nemitz, E.; Milford, C.; Sutton, M. A. A two-layer canopy compensation point model for describing bi-directional biosphere-atmosphere exchange of ammonia. *Q. J. R. Meteorol. Soc.* **2001**, *127*, 815–833.
- (75) Erisman, J. W.; Hensen, A.; Mosquera, J.; Sutton, M.; Fowler, D. Deposition monitoring networks: what monitoring is required to give reasonable estimates of ammonia/ammonium? *Environ. Pollut.* **2005**, *135*, 419–431.
- (76) Denmead, O. T.; Frenay, J. R.; Dunin, F. X. Gas exchange between plant canopies and the atmosphere: Case-studies for ammonia. *Atmos. Environ.* **2008**, *42*, 3394–3406.
- (77) Meng, Z.; Dabdub, D.; Seinfeld, J. H. Chemical coupling between atmospheric ozone and particulate matter. *Science* **1997**, *277*, 116–119.

ES8024403