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Measurements of ammonia emissions from oak and pine forests and development of a non-industrial ammonia emissions inventory in texas

Golam Sarwar[a,1], Richard L. Corsi[a,*], Kerry A. Kinney[a], Joel A. Banks[a], Vince M. Torres[a], Chuck Schmidt[b]

[a]*Center for Energy and Environmental Resources (R7100), The University of Texas at Austin, 10100 Burnet Road, Austin, TX 78758, USA*
[b]*Independent Environmental Consultant, 19200 Live Oak Road, Red Bluff, CA 96080, USA*

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Abstract

Estimates of non-industrial source ammonia emissions in Texas were developed through the use of published emission factors and activity data for those sources. A total of 64 non-industrial source emission sub-categories were addressed, each falling into one of seven major source categories: animal husbandry, fertilizer applications, on-road vehicles, non-road sources, municipal wastewater disposal, domestic sources, and natural soil and vegetation. Annual statewide ammonia emissions were initially estimated to be 921,000 metric tons, with greater than 50% originating from natural soil and vegetation. However, estimates for pine and oak forests were characterized as having a great deal of uncertainty. A series of field sampling events were conducted to determine ammonia fluxes from pine and oak forest floors in east Texas. Both dynamic and static chamber methods were used. The ammonia flux averaged 0.09 kg km^{-2} month^{-1} for pine forests and 0.13 kg km^{-2} month^{-1} for oak forests. These values are significantly lower than those previously measured and cited in the published literature. However, the ammonia fluxes measured in east Texas forests are reasonably consistent with those predicted using mechanistic models for evergreen pine and deciduous broadleaf forests in Alabama, California, Colorado, and Tennessee. Statewide annual ammonia emissions estimates, revised using the newly developed ammonia fluxes for oak and pine forests in Texas, dropped from 921,000 to 467,000 metric tons. The relative contribution of ammonia emissions from pine and oak forests dropped from 49% to less than 1%. Animal husbandry was predicted to be the dominant non-industrial source, accounting for approximately 77% of non-industrial source ammonia emissions.
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1. Introduction

Atmospheric chemical reactions are believed to be a major source of fine particulate matter ($PM_{2.5}$). An important contributor toward many of these reactions is ammonia (NH_3), which is emitted from a wide range of anthropogenic and natural sources.

*Corresponding author. Tel.: +1 512 475 8617; fax: +1 512 471 1720.
E-mail address: corsi@mail.utexas.edu (R.L. Corsi).
[1]Current address: USEPA, Mail Drop E243-03, 109 T.W. Alexander Drive, RTP, NC 27711, USA.

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Tropospheric concentrations of NH_3 are highly variable and dependent on proximity to sources, source strengths, meteorological conditions, and removal mechanisms. Typical atmospheric residence times for NH_3 are on the order of 10 days, and ammonia mixing ratios over continents generally range over two orders of magnitude, from 0.1 to 10 parts per billion by volume (ppb) (Seinfeld and Pandis, 1997). Dentener and Crutzen (1994) estimated global NH_3 emissions to be 45 million metric tons per year, with approximately two-thirds of this total being attributed to anthropogenic activities. Nearly one-half of global ammonia emissions were attributed to animal husbandry.

Chemical reactions involving NH_3 to produce secondary $PM_{2.5}$ depend on the presence and relative concentrations of atmospheric nitrates and sulfates. In areas characterized by high ammonia and nitric acid concentrations and low sulfate concentrations, gaseous ammonia can react to form ammonium nitrate. In the presence of sulfuric acid, increasing concentrations of gaseous ammonia can react to form ammonium sulfate. Whether reacted with nitrate or sulfate, the ammonium ion (NH_4^+) is often observed to be an important component of tropospheric aerosols. The conversion of ammonia to ammonium (NH_4^+) is also significant with respect to transport of NH_x, since the dry deposition of ammonia gas is generally 5–10 times faster than dry or wet deposition of ammonium-containing particles (Bouwman et al., 1997).

There is significant evidence that natural soil is an important contributor to global ammonia emissions (Dawson, 1977). For example, ammonium is found at relatively high concentrations in rainwater. Gaseous concentrations of ammonia are also greater over soils with high pH, a condition that shifts the acid-base equilibrium in soil from ammonium ion to ammonia. Atmospheric ammonia concentrations are greater over land than over oceans, and increase with increasing soil temperature. However, measurements of ammonia emissions from natural soils are sparse and corresponding emission factors are characterized by significant uncertainties. These facts are particularly true for ammonia emissions from forested areas, e.g., pine and oak forests that cover large areas of east Texas. The primary source of nitrogen that is converted to ammonia is organic nitrogen associated with foliar litter. Thus, greater amounts of fresh litter deposition should lead to increased ammonia emissions.

The intent of the study described herein was to develop a first estimate of non-point source ammonia emissions in Texas. A total of 64 non-point sources of ammonia were considered in this study. Each source required significant reviews of existing literature and relevant databases prior to the selection of appropriate emission factors and source activity data. Given the extensive nature of these tasks, it is impossible to describe all aspects of the study in this paper. Instead, we have described the project methodology in general terms, and have listed several important references and databases. The reader is referred to the complete project report for a more extensive description of methodologies and results (Corsi et al., 2000a). We do provide details related to actual ammonia flux measurements from forest floors in east Texas and use the results to facilitate the overall ammonia inventory.

2. Methodology

An extensive literature review was completed in order to identify potential non-industrial sources of ammonia emissions; As well as to identify and assess relevant emission factors. Ten bibliographic databases were searched using "ammonia" and "emissions" (inclusive) as keywords. Forty web sites were also found to contain information related to ammonia emissions, 14 of which were identified as relevant to this project. Personal contacts were also made with individuals known to be, or who are known to have been, involved with ammonia emissions estimates. In total, 655 publications were identified as containing information relevant to this project. Approximately 120 of these publications were selected for thorough review. Through this process, it quickly became evident that a small number of previous publications are frequently referenced and used by others to estimate ammonia emissions (Asman, 1992; Battye et al., 1994; Bouwman et al., 1997; Buijsman et al., 1987; Gharib and Cass, 1984; Klaassen, 1991; and Lee and Dollard, 1994).

2.1. Selection of source categories

Based on a review of existing literature, a total of seven non-industrial source emission categories were selected for this study. Sixty-four sub-categories that fall within the major source categories are listed in Table 1. While it was obvious at the beginning of this study that some sources would be relatively insignificant, e.g., rabbits and untreated human waste, for completeness emission factors and

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