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Ammonia emissions from biomass burning in the continental United States

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ABSTRACT

This study quantifies ammonia (NH_3) emissions from biomass burning from 2005 to 2015 across the continental US (CONUS) and compares emissions from biomass burning with the US Environmental Protection Agency (EPA) National Emissions Inventory (NEI), the Fire Inventory from the National Center for Atmospheric Research (FINN) and the Global Fire Emissions Database (GFED). A statistical regression model was developed in order to predict NH_3 emissions from biomass burning using a combination of fire properties and meteorological data. Satellite data were used to evaluate the annual fire strength and frequency as well as to calculate the total NH_3 emissions across the CONUS. The results of this study showed the total fire number has decreased, while the total yearly burn area and the average fire radiative power has increased. The average annual NH_3 emissions from biomass burning from this study, on a national scale, were approximately 5.4e8 ± 3.3e8 kg $year^{-1}$. When comparing the results of this study with other emission inventories, it was found that ammonia emissions estimated by the NEI were approximately a factor of 1.3 lower than what was calculated in this study and a factor of 1.1 lower than what was modeled using the statistical regression model for 2010–2014. The calculated NH_3 emissions from biomass burning were a factor of 5.9 and a factor of 13.1 higher than the emissions from FINN and the GFED, respectively. The modeled NH_3 emissions from biomass burning were a factor of 5.0 and a factor of 11.1 higher than the emissions from FINN and the GFED, respectively. As the climate continues to change, the pattern (frequency, intensity and magnitude) of fires across the US will also change, leading to changes in NH_3 emissions. The statistical regression model developed in this study will allow prediction of NH_3 emissions associated with climate change.

1. Introduction

Ammonia (NH_3) is an important base gas in the atmosphere (Battye et al., 2017; Aneja et al., 1998, 2008). NH_3 reacts with sulfuric, nitric and hydrochloric acids to form ammonium sulfate, ammonium bisulfate, ammonium nitrate and ammonium chloride which contribute to the formation of $PM_{2.5}$ (particulate matter with diameter less than 2.5 micrometers) (Baek and Aneja, 2004; Baek et al., 2004; Davidson et al., 2011; Day et al., 2012; Chen et al., 2014). There are many adverse health effects associated with exposure to elevated concentrations of fine particulate matter, such as chronic bronchitis, aggravated asthma, irregular heartbeat, other cardiovascular and respiratory issues and even death (Pope et al., 2002, 2009; Schwartz et al., 2002; Kwok et al., 2013; Crouse et al., 2015; Lelieveld et al., 2015). Exposure to elevated $PM_{2.5}$ concentrations is a major concern for human health and welfare due to the particles' ability to penetrate deep into the respiratory tract. $PM_{2.5}$ is also associated with several environmental impacts, such as reducing visibility and changing the earth's radiational balance (Fan et al., 2005; Behera and Sharma, 2010a, 2010b; Heald et al., 2012; Wang et al., 2012). Furthermore, gaseous NH_3 may be deposited to the Earth's surface, which leads to ammonification, eutrophication and a loss of biodiversity (Langford et al., 1992; Robarge et al., 2002; Galloway et al., 2004; Clark and Tilman, 2008; Janssens et al., 2010; Day et al., 2012; Holtgrieve et al., 2011; Phoenix et al., 2012; Erisman et al., 2013; Chen et al., 2014). Increased concentrations of NH_3 can also lead to a decreased resistance to drought and frost damage (Robarge et al., 2002). In addition, NH_3 plays a role in the formation of nitrous oxide, which is a major greenhouse gas (Bouwman, 1996).

Major sources of atmospheric NH_3 include NH_3 based fertilizers, animal waste, and biomass burning, with intensely managed livestock and agricultural sources of NH_3 contributing most to NH_3 concentrations (Langford et al., 1992; Schlesinger and Hartley, 1992; Bouwman et al., 1997; Flechard and Fowler, 1998; Battye et al., 2003; Aneja et al.,

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2009; Zbieranowski and Aherne, 2012). While agriculture accounts for approximately 82% of all NH_3 emissions on a national level, fires account for a total of about 10% of all ammonia emissions nationwide (2014 NEI). NH_3 is mainly emitted into the atmosphere during smouldering combustion, which occurs in slow, low-temperature fires without a flame (Langford et al., 1992; Nance et al., 1993; Goode et al., 2000; McMeeking et al., 2009; Akagi et al., 2011; Alves et al., 2011; Chen et al., 2014).

Previous works have shown an overall increase in NH_3 sources (Erisman et al., 2008) and atmospheric NH_3 concentrations over the past several years (Saylor et al., 2015; Butler et al., 2016; Yao and Zhang, 2016). While the overall increase in NH_3 emissions cannot solely be attribute to fire activity, it is possible that biomass burning emissions of NH_3 are contributing to the observed increase in ambient emissions. For example, Saylor et al. (2015) also observed unusually high concentrations of NH_3 across the southeastern United States (US) during 2007, when fires were prevalent due to increased temperatures and widespread drought. Similarly, R'Honi et al. (2013) observed NH_3 concentrations were two orders of magnitude larger than background levels during the summer of 2010, which was the hottest and driest summer on record (until 2015), when wildfires ran rampant across Europe and Russia. Hot and dry conditions in the Mediterranean countries, Australia and the western United States have contributed to an increase in wildfire activity, thus increasing the emission of gaseous NH_3, among other pollutants, into the atmosphere (Alves et al., 2011). The strength and frequency of fires are not only controlled by the properties of the fuel and the geography, but they are also influenced by weather and climate (Pyne et al., 1996; Liu et al., 2010). Therefore, changes in the earth's climate will likely result in changes in fire activity. Higher temperatures and widespread drought are expected to cause an increase in the number of observed wildfires across many regions, such as the southeastern United States, the northern great plains, the Pacific coast, the southwestern US and the southern Rockies (Piñol et al., 1998; Gillett et al., 2004; Reinhard et al., 2005; Liu, 2006; Westerling et al., 2006; Alves et al., 2011; Litschert et al., 2012; Saylor et al., 2015; Skibba, 2015). However, due to changes in relative humidity and wind speeds, the future fire potential in the northern Rockies and the northwestern United States may likely be reduced (Liu et al., 2013). On a global scale, wildfire potential is projected to increase as the climate changes, specifically in locations that are already prone to the occurrence of wildfires (Liu et al., 2010). This increase in wildfire potential will then potentially lead to an increase in NH_3 emissions from biomass burning.

Biomass burning is an important source of NH_3 emissions, but the strength of the source remains poorly quantified (Alves et al., 2011; Chen et al., 2014). Therefore, the primary objective of this study is to quantify NH_3 emissions from biomass burning (wildfires, agricultural burns and prescribed burns) from 2005 to 2015 across the continental US and compare against major emission inventories used in atmospheric models. The inventories compared in this study include the Fire Inventory from the National Center for Atmospheric Research (FINN v1.5, Wiedinmyer et al., 2011), the Global Fire Emissions Databases (GFED v4.1, with small fires; Van Der Werf et al., 2017), and the US Environmental Protection Agency (EPA) National Emissions Inventory (NEI). As described in Larkin et al. (2014), the US EPA NEI is produced every three years and includes state submitted data. For this study, the years 2011 and 2014 are NEI process years while the remaining years in this study are considered fire inventory data, which were compiled using a more limited set of inputs. Therefore, while the US EPA emissions data is referred to as NEI in this study, it is important to remember that only 2005, 2008, 2011 and 2014 are NEI process years and the remaining years are based on EPA fire inventory data. Furthermore, a regression analysis, using forward stepwise regression, was completed in order to determine the best fitting model of NH_3 emissions from biomass burning using a combination of in-situ and satellite (primarily NASA's Terra and Aqua) observations. This work proposes a new methodology to project emissions of NH_3 on a national scale, which would help society understand the implications of the changing climate and adequately prepare and/or prevent these changes. Furthermore, this methodology also provides a relatively simply approach to estimating past, present and future emissions based on readily accessible data (temperature and burn area).

2. Data & methodology

In order to compare the calculated fire emissions (discussed in the following section) with the fire properties (number of fires, fire radiative power, and fire brightness temperature) as well as to observe trends in the fire properties, the National Aeronautics and Space Administration's (NASA) Fire Information for Resource Management System (FIRMS) was utilized to obtain archived fire locations, frequency and strength. This data was obtained from the MODIS sensor on NASA's Earth Observing System satellites (Terra and Aqua) (Friedl et al., 2010). The MODIS active fire product obtained (Collection 6, Giglio et al., 2016) uses a fire detection algorithm that uses a multispectral contextual approach to leverage the mid-infrared radiations emitted by fires (Davies et al., 2009). FIRMS delivers the MODIS fire data locations that represent the center of a 1 km pixel that is flagged by the algorithm as an area that contains at least one fire/hotspot within the pixel (Davies et al., 2009). The brightness temperature is calculated using the average intensity of infrared radiation at two wavelengths near 4 μm for a 1 km × 1 km pixel (Giglio et al, 2003, 2016). In the Collection 6 MODIS active fire product, the fire radiative power was derived using the Wooster et al. (2003), Wooster et al. (2012) approach (Giglio et al., 2016). In order to ensure quality, only fire data with a confidence estimate greater than 33% (i.e. medium and high confidence fires) will be used in this study.

2.1. Quantification of NH_3 emissions

There are several methods that can be used to quantify emissions of pollutants (i.e. NH_3) from biomass burning (i.e. Van Der Werf et al., 2003; Hoelzemann et al., 2004; Ito and Penner, 2004; Van Der Werf et al., 2014; Dennis et al., 2002; Langmann et al., 2009; Ichoku and Ellison, 2014). In this study, ammonia emissions from biomass burning were calculated using the emission factor approach (Equation (1)) equation adapted from Seiler and Crutzen (1980), Wiedinmyer et al. (2006), Wiedinmyer et al. (2011) and Oliveras et al. (2014):

$$E_i = BA_{(x,t)} \times B_{(x)} \times FB \times EF_j \quad (1)$$

where E_i is the emission of species i (in this case, NH_3), BA is the area burned at time t and location x, B is the biomass loading at location x, FB is the fraction of that biomass burned in the fire and EF_i is the emission factor of species i. In order to obtain the area burned (BA), the Moderate Resolution Imaging Spectroradiometer (MODIS) Burned Area product (MCD45, Collection 5.1), obtained from the University of Maryland's website, was used. The burn area is determined by the MODIS algorithm that uses time series of the daily 500 m MODIS land surface reflectance data (Roy et al., 2002, 2005, 2008). The MODIS burned area product was validated by Roy et al. (2005) and then again by Roy and Boschetti (2009), who found that the MODIS product provided the most accurate burned area maps when compared with other products (i.e. L3JRC, GlobCarbon). The biomass loading (B), which is defined as the amount of biomass available that can be burned in each fire, was obtained from Table 1 in Wiedinmyer et al. (2006), which describes the total fuel loading assumptions for various land cover classifications based on the literature. In order to quantify the amount of biomass burned, it was first necessary to know the type of land being burned. Therefore, the Collection 5 MODIS Global Land Cover Type product for 2010 (MCD12Q1) was used to determine land type (Friedl et al., 2010; Channan et al., 2014). This database contains land cover classifications at a spatial resolution of 500 m. This data was readily