Inhibition of Soil Methane Oxidation by Fertilizer Application: an Intriguing but Persistent Paradigm

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Abstract. Methane (CH₄) is one of the most important greenhouse gases and is oxidized by the methanotrophic bacteria in the soil. Present work is an effort to review the available information in this regard and present them in a systematic way. In this review, we concluded that low NH₄⁺ concentration can be supportive to the methane oxidation and growth of the methanotrophs. However, their high contents suppress the methanotrophic bacteria by inhibiting the enzymes particularly methane monooxygenase (MMO) involved in the methane oxidation. There are a range of the soil and environmental factors such as type of soil and vegetation, methane availability, amount and exposure time of ammonium, and type of methanotrophic community dominating in an ecosystem, which affect the response of the methanotrophic bacteria towards the fertilizer application. However, still there are several gaps in our knowledge as complex interaction of edaphic factors affecting the availability of ammonium is unraveled.

Key Words: Methane, soil methane oxidation, fertilizer application, ammonium fertilizer, nitrate fertilizer, organic fertilizer

1 Introduction

Methane (CH₄) is a radiatively active hydrocarbon present in the atmosphere. Its global warming potential is 21, which makes it responsible for approximately 15% of the total greenhouse effects [1], [2], [3]. The estimates of annual methane emission were around 300 Tg during 2000 which was projected to increase up to 400 to 600 Tg in 2010 [4]. Global methane emission is balanced by the atmospheric and soil methane sink activity. Atmospheric sink activity is responsible for the 90% of the global methane sink capacity, while rest 10% is mediated by methane oxidation capacity of the soil. Soil methane oxidation is carried out by the aerobic eubacteria called methanotrophic bacteria (MB) or methane oxidizing bacteria (MOB). Growth of methanotrophs is limited to CH₄, though some may utilize methanol and in some other cases formate, formaldehyde, methylamine etc. [5]. Methanotrophs oxidize methane in the presence of the enzyme methane monooxygenase (MMO). They are divided into Type I, Type II, and Type X based on their intra membrane pattern, physiological characteristics, chemotaxonomic nature and phylogenetic position [6]. However, recently it has been suggested that they should be categorized into only Type I and Type II. Type I methanotrophs (∝-proteobacteria) include Methylomonas, Methylobacter, Methylosarcina, Methylocaldum, Methylococcus, and Methylocapsa, while Type II methanotrophs (∝-Proteobacteria) include Methylosinus, Methylocystis, Methylocella and Methylocapsa. A range of natural as well as anthropogenic factors influence the methane oxidation potential of the soil. Among them, effect of the fertilizer application on the methane oxidation remains one of the most studied aspects. Nitrogenous fertilizers are generally thought to have an important role in regulating methane oxidation. In this context, the effect of ammonium on methane oxidation activity in different soil types is an important aspect and has been instigated by several works. The inhibition of methane consumption potential of soils by mineral fertilizer was firstly reported in temperate forest soil [7]. Since then, a range of ecosystems have been studied to assess the consequences of fertilizer application which have given contradictory reports depending upon the nature and amount of fertilizer, longevity of the treatment, type of ecosystem, and edaphic factors prevailing in a particular ecosystem.
There are several reports suggesting the inhibition of the methane oxidation capacity of the soil by the application of the mineral fertilizers. The subject is of high relevance, yet no systematic review is available in this context. The present work is an effort to review the available information in this regard and present them in a systematic way. It is noteworthy that the interactions between the nitrogen and methane cycle are complex and not completely understood. Several studies were performed to investigate the effects of N fertilizers on methane oxidation to better understand the interactions between the methanotrophic community and ammonia oxidizing bacteria. Under this review we tried to develop a complete picture of this process which is delicately balanced by nature. The accounts of methane consumption potential of the soil as affected by the different fertilizers (Table 1) and possible operative mechanisms have been discussed in coming sections.

2 Ammonium Fertilizers

Out of the total fertilizer consumption, share of nitrogenous fertilizers is 80-90% of which ammonium fertilizers account for 70% [9]. Ammonium application has shown conflicting results in relation to inhibition of soil methane oxidation [10], [11] stimulation [12] or even no effect [13], [14] of CH₄ oxidation, consumption or sink activity which may vary from upland (e.g., forest and grassland) to lowland soils (e.g., rice paddies and fresh water marshes) [15],[16]. Longevity of NH₄⁺ application also affected the response of methanotrophic bacteria. In short term, NH₄⁺ was either inhibitory [17] or exerted no effect [18] while in long term, it usually showed inhibitory effect. [18]. The inhibition of NH₄⁺ still occurred even when NH₄⁺ concentration has decreased to the background levels [19],[20],[21]. Some scientists have observed that inhibitory effect decreased after long term NH₄⁺ application [17],[22],[23]. It might be due to increased formation of organic C by N application resulting into the immobilization of excess NH₄⁺-N. Similarly, inhibitory effect was found to disappear after yearly application of fertilizer [24] probably, due to adaptation in methanotrophic population against the elevated ammonium concentration in long term. Longevity of inhibition was also dependent upon the endogenous level of the NH₄⁺ in soils as CH₄ uptake activity of the forest soils characterized by N limitation recovered rapidly from the inhibitory effect [25]. Level of the ammonium added may also play a critical role in determining the response of the soil. At low NH₄⁺ concentration, CH₄ oxidation activity was unaffected, while at higher concentration CH₄ oxidation was significantly reduced. Any factor ensuring the persistent but slow release of the N usually enhanced methane uptake. Mori et al. observed that nitrogen fixed by leguminous plants did not reduce the CH₄ uptake probably due to long term sustained and slow release of NH₄⁺ in soil [26].

3 Factors Affecting Soil Methane Oxidation

A range of the edaphic and natural factors affect the level of reactive and available NH₄⁺ in a particular ecosystem. The effect of ammonium is controlled by CH₄ concentration in the environment. In non-water saturated soils, where CH₄ uptake took place at low CH₄ concentrations, increased NH₄⁺-availability usually suppressed CH₄ oxidation rate [7],[27],[28],[29],[30]. While, in water saturated wetland soils characterized by high CH₄ concentration, the effect was contradictory, with reports ranging from inhibition to stimulation [31],[32],[33],[34],[35]. The initial inhibitory effect of NH₄⁺ was eliminated during subsequent incubations under high CH₄ concentration (>1000 ppm), while the effect was persistent at low methane concentration (<500 ppm) [35]. In its agreement, it has been reported that N fertilization suppressed CH₄ oxidation in surface soil, while having no effect in subsurface soil [36],[37]. Higher methane concentration through endogenous methane production in subsurface layer may be responsible for no effect of fertilizer on methane oxidation in subsurface soil.

Besides, soil types have also been reported to significantly affect the relation between ammonium and CH₄ oxidation rate. Clay soil binds positively charged NH₄⁺ strongly, thus preventing its leaching and ensuring long term availability, while at the same time in sandy soil it would be readily available to negatively affect the methane oxidation. Tanthachoom et al. noticed that the effect of NH₄⁺ was inhibitory in sandy loam soil, while at the same time, stimulatory to some extent in compost soil which is characterized by low pH and high humus content. In soil, rich in organic C, excess ammonium is immobilized, thus releasing the NH₄⁺ slowly but persistently affecting the methane oxidation favorably.
NH₄Cl caused greater inhibition compared to NH₄(SO₄)₂ presumably due to increased ammonium and increasing the extent and amount of atmospheric acid deposition. Effect of ammonium was also connected with type of forest soil. Rigler and Zechmeister-Boltenstern reported that NH₄ desorption of ammonium. It was evidenced by the observations of King and Schnell, who reported that the oxidation of ammonium [45]. Direct evidence for nitrification by methanotrophs shows in [48]. Non ammonical salts were having varying effect on the availability of ammonium [41]. Different anions were having varying effect on desorption of ammonium. It was evidenced by the observations of King and Schnell, who reported that NH₄Cl caused greater inhibition compared to NH₄(SO₄)₂ presumably due to increased ammonium desorption by the sulphate. [41]Likewise, the load of the non ammonical salts may also influence the desorption potential of the ammonium methane oxidation differently.[41] Non ammonical salts were found as essential controls for partitioning ammonium inhibition between the nonspecific and MMO related mechanisms [42], [43].

4 Mechanisms for the Inhibition of Methane Oxidation by Ammonium Fertilizers

Based on findings of different studies an explanatory hypothesis has been proposed (Fig. 1) to explain the mechanism of inhibition of methane oxidation by ammonium fertilizers. Among them, the most widely held theory is that MMO enzymes is able to oxidize NH₄⁺ to NO₂⁻ also, along with oxidizing methane to methanol [44], [45], [46], [47] and therefore NH₄⁺ acts as a competitive inhibitor of MMO [48], [45]. Whereas, Dunfield and Knowles, could not detect whether this competition is simple or partial [36]. However, according to them simple competition inhibition is more likely. It is due to high level of similarity between pMMO (particulate methane monoxygenase) found in methanotrophs and AMO (ammonia monoxygenase) enzyme found in ammonium oxidizing bacteria [49]. These similarities include a high degree of amino acid sequence identity, similar protein complex structures, similar substrate and broadly inhibition profiles [50], [51], [52]. Methanotrophs are unable to obtain energy from the oxidation of ammonium [45]. Direct evidence for nitrification by methanotrophs shows in [48]. Yoshinari et al, has been given only for pure cultures. In most of the natural ecosystems, it has been demonstrated only indirectly by means of the inhibitor-sensitive ¹⁴CH₄/¹⁴CO oxidation ratio, which is higher for methanotrophs than for nitrifiers [53], [54]. Several inhibitors (e.g., C₃H₇, CH₃F, dimethylether, allylsulfide, allylthiourea, dicyandiamide, picolinic acid, and difluoromethane) have been evaluated for their potential to selectively knock out one group of bacteria without affecting the other [55]. However, only allyl sulfide [56] and picolinic acid [57] showed potential for discrimination, although neither was able to discriminate 100%. O’Neill and Wilkinson [48], suggested that nitrite inhibits the formate dehydrogenase, an enzyme producing reductant for CH₄ oxidation. The hydroxylamine (NH₂OH) and nitrite (NO₂⁻) produced by NH₄⁺ oxidation was also suggested to suppress methanotrophic activity [29], [58]. There are reports indicating that NH₄⁺ as well as high concentration of NH₃ inhibits another key enzyme of methane cycle, methanol dehydrogenase (MDH) converting methanol to formaldehyde. In its agreement, Boiesen and Arvin reported that ammonia is a weak competitive inhibitor as observed by other researchers and relatively high levels ammonia (70 mg/l) was able to inhibit the methane consumption [59]. Addition of NH₄⁺ fertilizers may also stimulate the ammonium oxidizing bacteria (AMO) which may occupy niche of certain methanotrophic bacteria resulting into reduced CH₄ oxidation [60], [61],[62]. However, their population size is not significantly higher than MOB in soil [63] and the rate of CH₄ oxidation carried out by them is about 100-10,000 times less than methanotrophs.
Henckel et al., [65] could not detect this group of bacteria in the surface forest soil through amplification of the amoA gene of ammonium oxidizers. So probably they are not the candidates responsible for atmospheric methane uptake in soil [66]. Similarly, several other studies including those of the Bodelier and Frenzel conducted in rice field and Henckel et al., in forest soil revealed insignificant contribution of nitrifying population in methane oxidation [55], [65]. 14C PLFA analysis also excluded any significant role of ammonium oxidizers in the metabolism of atmospheric CH4 [67]. Positive relation between ammonium and methane oxidation at higher methane concentration is resulted from the inability of ammonium to competitively inhibit the MMO, while at same time supplying requisite level of nutritional N. Another explanation may be that nitrifying bacteria are stimulated under high ammonium concentration, which in turn oxidizes CH4 under elevated CH4 concentration [62], [68]. Work of Dunfield and Knowles [36] suggests that instead of NH4+, NO2− is more responsible for the inhibition of the methane uptake and production of the NO2− from the NH4+ is diminished under high methane concentration. However, few studies have reported the inhibition of methane oxidation or stimulation of the NH4 oxidation even at higher methane concentration[29] [46]. It is presumably due to alleviation of NADH limitation [36]. Another study by Gulledge and Schimel concluded that the increased inhibition of CH4 oxidation by mineral N under higher methane as observed by King and Schnell was due to Cl−, the counter-ion of NH4+ in their study [29], [69]. Whalen and Reeburgh also expressed same view [37]. However, crucial role of Cl− as suggested by these workers was contradicted by King and Schnell, who reported that (NH4)2SO4 also inhibited the methane oxidation under high methane concentration [41].

Besides, NH4+ can have non-specific inhibition mechanisms which may include an inhibitory salt effect (osmotic stress) and decrease in soil pH [69], [41], [36]. Former, assumption has been proposed due to the inhibition brought about by several other nitrogenous and non-nitrogenous salts also [70], [41], [69], [71]. It was also ensured by the conductivity measurement of the soil after salt addition [72]. Saari et al., [72] observed that methane oxidising microbes were more sensitive to the salt treatment than other soil microbes based on the CO2 production rates. Further, they suggested that pH variation following the ammonium amendments have very little effect if any, contrary to the earlier reports. In addition to the

![Figure 1. Enzymatic inhibition of the methanotrophic bacteria by the ammonium and derived products](image-url)
above mechanisms, nitrogen addition may increase root biomass in forest soil resulting into decreased soil porosity and restricted gas diffusion hindering the CH$_4$ oxidation [73]. Steudler et al. [74] suggested that the direct impact of ammonium is related to its inhibitory effect on the CH$_4$ oxidizing bacteria, while its indirect effect is caused by change in nitrogen turnover rates. Other studies also suggested that the N turnover rate rather than the mineral N content influences the CH$_4$ oxidation. [19] Evidently, low nitrogen turnover is responsible for high CH$_4$ oxidation rate in forest soil [75],[64],[76].

Net effect of the ammonium on the methane sink activity of the soil (Fig. 2) should be dependent on the response of both the methanotrophs as well as methanogens to fertilizer additions. However, related studies have been seldom carried out in any ecosystem. Some idea can be generated by the work of Zhang et al. in the soils of submerged wetland [12]. They have suggested three effects of ammonium application on methanogens stimulating plant growth and therefore intensifying CH$_4$ emission by providing more methanogenic substrates or improving aerenchyma conditions, and intensifying CH$_4$ oxidation by providing O$_2$ to the rhizosphere due to improvement of aerenchyma conduits and accordingly decreasing CH$_4$ emission.

![Figure 2. Proposed mechanisms for inhibition of the methane consumption](image)

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5 Ammonium Fertilizers and Methanotrophic Community

Ammonium exerts its effect at the community level also. Enumeration of methanotrophic population size through MPN (most probable number) technique showed that ammonium application decreased the methanotrophic population size [77],[10]. Molecular and biochemical techniques have also yielded similar results. Seghers et al, observed that fatty acids characteristics for methanotrophs were less abundant in the soil treated with mineral fertilizer compared to that treated with compost [78]. It has been suggested that differential responses of soil subsequent to fertilizer application may be due to different dominating methanotrophic communities in the soil [60], [79], [80]. PLFA (phospholipid fatty acid analysis) along with SIP (stable isotope probing) revealed that application of NH$_4^+$ in presence of high CH$_4$ concentration reduced the amount of $^{13}$C incorporated into majority of PLFA except few, indicating that suppression of one group of methanotrophs occurred more readily than others [81]. Type I methanotrophs are stimulated under the presence of elevated NH$_4^+$ concentration contrary to Type II, possibly due to N$_2$ fixing ability of latter [34],[80],[82],[83],[84]. Lau et al., [84] performed rRNA targeted quantitative hybridization in the pine forest soil and suggested that Type II were more sensitive to nitrogen addition. However, all the Type II methanotrophs are not prone to the NH$_4^+$ addition [84]. Noll
et al., [85] observed that exogenous NH$_4^+$ application in rice field soil activated only a small subset belonging to *Methylobacterium/Methylocaldum* within Type II methanotrophs. Both low as well as high affinity methanotrophs are adversely affected by the NH$_4^+$ treatment [86].

6 Effect of Nitrite

Applications of NO$_2^-$ resulted into inhibition of methane oxidation soils and sediments incubated in laboratory conditions found that exogenous NO$_2^-$ was a more effective inhibitor of CH$_4$ consumption than NH$_4^+$ in an acid forest soil (59% and 42% inhibition, respectively) [29], [87]. In forest soil, the inhibitory effect of NO$_2^-$ was shown to be greater and more enduring than the direct effect of NH$_4^+$. Dunfield and Knowles [36] observed that addition of NO$_2^-$ (40 mg N kg$^{-1}$ soil) inhibited CH$_4$ oxidation by 84% initially, which decreased up to 41% after 48h but its negative effect persisted till it was completely converted into NO$_3^-$. Same workers reported nitrite has minimal secondary toxic effects relative to direct inhibition caused by ammonium. The relative inhibition caused by NO$_2^-$ was highest at low CH$_4$ concentrations however, its production through nitrification by methanotrophic culture increased with methane concentration [29], [36]. Nitrite is a non-competitive inhibitor of methane oxidation. It can inhibit formate dehydrogenase and can contribute to NADH limitation, explaining its greater influence on CH$_4$ limited cells [29], [36], [88]. A high metabolic rate might also be necessary to export toxic, cellular NO$_2^-$ produced by the nitrification in methanotrophs [36]. It was found that NO$_2^-$ concentrations produced through nitrification of the ammonium were lower than required for inhibition of methane oxidation, but its intracellular concentration may be higher than required, due to which methanotrophs might produce NO$_2^-$ themselves [36]. Production of NO$_2^-$ from NH$_4^+$ in methanotrophs cultures increased with increasing CH$_4$ concentrations [87] further implicating NO$_2^-$ as a significant inhibitor, as previously noted [29], [36], [89].

Perhaps the absolute concentration and time of exposure affect the ability of methanotrophs to recover from NO$_2^-$ inhibition and perhaps the extremely high natural nitrification rate of this humisol shields methanotrophs from NO$_2^{-}$ [36]. Hutsch suggested that in soils with optimal conditions for nitrification, the inhibitory effect of NH$_4^+$ via NO$_2^-$ is unlikely as it is immediately oxidized as soon as it is produced. Therefore, the observed 64% inhibition of CH$_4$ oxidation after nitrification of the added N could have partly resulted from the concurrent drop in soil pH [90].

7 Effects of Nitrate Fertilizers

Similar to the ammonium, there are conflicting results about the influence of nitrate addition on the methane oxidation activity of the soil. Several workers have reported that there was no effect of nitrate (NO$_3^-$) on methane consumption [28], [29], and [38], while other workers have suggested that its effect was dose dependent. Lower concentration of nitrate has neither effect on methane consumption such as in deciduous forest soil, nor stimulatory effect such as in spruce forest soil; however, at higher concentrations, it suppressed the methane consumption at both sites [15]. In case of inhibition caused by the nitrate, the extent and duration was usually less compared to ammonium [29], [88], [92], [93]. However, an effect more acute than was caused by ammonium common. After nitrate addition, 10 to 86% reduction in the CH$_4$ oxidation rate was observed in forest soils [3]. Studies on pure cultures of CH$_4$ oxidizers have also suggested a direct inhibitory effect of nitrate on methanotrophs [3]. It may be due to toxic effect of NO$_3^-$ itself or NO$_2^-$ produced via NO$_3^-$ reduction. However, the exact mechanism for this inhibition is still unknown. Wang and Ineson suggested that cations associated with nitrate rather than nitrate itself are responsible for producing the inhibitory effect. Like ammonium, nitrate also affects both low and high affinity methanotrophs [46]. However, effect was direct on low affinity methanotrophs but indirect on high affinity methanotrophs [86], [93].

8 Effects of Organic Fertilizers

Organic fertilizer treatments have either no effect on CH$_4$ oxidation rate or inductive effect [18], [94]. But effect was negative when added in combination with mineral fertilizer, this may be due to immobilization of added mineral N and their regular release thereafter [94]. Both low and high affinity
methanotrophs are stimulated by the application of the organic fertilizer [86]. Organic fertilizer induced some particular group of methanotrophs different from those induced by mineral fertilizer. It was evidenced by DGGE (denaturing gradient gel electrophoresis) analysis of 16s rRNA gene fragment specific for methanotrophs which revealed a distinct community between mineral and organic fertilized soils as extra Type I methanotrophic bands became visible in the organic fertilized soils [95],[96].

9 Global Implications and Future Perspective

Methane source capacity of the soil is dependent upon the methane consumption potential of the soil which is ultimately dependent upon the several natural and anthropogenic factors prevailing in a particular ecosystem. The matter has become more complex by the effect inflicted by the fertilizer on methane consumption [97],[98],[99],[100]. The accounts of methane consumption potential of the soil as affected by the different fertilizers are given in table 1 and under this we try to discuss various operative mechanisms. Application of the fertilizer has increased to elevate crop production and feed the ever-increasing human population particularly since green revolution started in Asian countries during 1950s. Further, for several years developed countries have resorted to fertilizing the natural forest usually characterized by the N limitation, with the aim of increasing forest productivity (Fig.3). World wide consumption of nitrogenous fertilizer increased from 13 mt (million tones) in 1962 to 97 mt in 2006, which is projected to reach up to 118 mt in 2011, [9]. This increase is likely to be caused by the high domestic demand in the Asian countries which was responsible for 62% of the total global fertilizer consumption [9]. Besides, these regions harbor vast rice fields (90% of the total world) and large cattle population. The cumulative effect of both the factors is likely to affect the share of these countries in global green house gas emission. Due to obligations set under the Kyoto protocol to the different countries for reducing the emission of green house gas it is important to precisely determine the methane source capacity of different regions on the globe. It will require the effect of the nitrogenous fertilizers on the methane oxidation activity.

Figure 3. Regression analysis between (a) yearly global fertilizer consumption and methane emission and (b) country wise fertilizer consumption and methane emission

From the above discussion, it is clear that response of methane oxidation and methanotrophic bacteria towards the nitrogenous fertilizers depends upon several factors such as soil type, type of vegetation, methane availability, amount and exposure time of ammonium and type of methanotrophic.
community dominating in a ecosystem. However, the story is still not complete and there are many gaps in knowledge regarding the response of this bacterial group to fertilizer amendments. The complex interactions between the various natural and anthropogenic factors that affect the availability of ammonium for the MOB population have to be explored. Further, community level investigation has not been carried out to find out the response of high affinity methanotrophic community; whether the community size shrink or shift in response to elevated NH$_4^+$ concentration is not fully understood. Since inhibition of methane oxidation will increase the atmospheric methane load and increased green house affect, we can safely recommend that instead of conventional fertilizers we should use slow release fertilizer in agroecosystems. We should again resort to organic fertilizers which will alleviate the negative effect of the mineral fertilizer as well as decrease the methane emission from the stored organic manures by creating aerobic condition.

**Table 1.** Inhibitory effects of inorganic nitrogen on methane oxidation in various forest soils

| Study site                        | Nitrogen (concentration)                                                                 | CH$_4$ concentration | Effect         | References |
|----------------------------------|-----------------------------------------------------------------------------------------|----------------------|----------------|------------|
| Pure culture                     | NH$_4$Cl (>0.05 % (w/v))                                                              | >1 to 10mM           | 5% reduction  | [97]       |
| Taiga forest                     | 200 Kg Nha$^{-1}$$^{1}$                                                                |                      |                |            |
| Canada coniferous forest          | (NH$_4$)$_2$SO$_4$ (5 and 10 mM)                                                        | 10 ppm               | 61-95% reduction | [75]       |
|                                   | NH$_4$Cl (10 mM)                                                                       |                      | 93% reduction  |            |
|                                   | NaNO$_3$ (10 mM)                                                                       |                      | 75% reduction  |            |
| Pure culture of methanotroph      | NH$_4$Cl (0-200µM)                                                                     | 100ppm               | 20-75%         | [29]       |
|                                   | NH$_4$Cl (500µM)                                                                       | 1.7 to 1000 ppm      | 40-100%        |            |
| Scot pine forest, Norway          | NH$_4$NO$_3$ (30 and 90Kg ha$^{-2}$ a$^{-1}$)                                           | Ambient air           | 85 and 62% reduction | [86]       |
| Scotland deciduous forest         | NH$_4$NO$_3$ (150 or 226 kg N ha$^{-1}$)                                               | Ambient air           | reduction      | [11]       |
| Taiga forest, USA,                | NH$_4$NO$_3$ (60 and 50 kg N ha$^{-1}$ a$^{-1}$)                                       | Ambient air           | 0-75% reduction | [60]       |
| Scotland coniferous forest        | NH$_4$NO$_3$, NaNO$_3$, NH$_4$Cl: 40 kg N ha$^{-1}$                                    | Ambient air           | NH$_4$NO$_3$: 87% | [92]       |
|                                   |                                                                                       |                      | Na$_2$CO$_3$: 86% NaCl: 70% |            |
| Maine forest soil, Germany        | NH$_4$Cl (1mM g soil$^{-1}$)                                                           | 1.7ppm               | 99.5% reduction | [41]       |
|                                   | (NH$_4$)$_2$SO$_4$ (1mM g soil$^{-1}$)                                                 | 270ppm               | 60% reduction  |            |
|                                   |                                                                                       | 1.7ppm               | 99.5% reduction |            |
|                                   |                                                                                       | 270ppm               | 96% reduction  |            |
| Arable soil, UK                   | (NH$_4$)$_2$SO$_4$ (48, 98, 192 kg N ha$^{-1}$)                                        | Ambient air           | reduction      | [99]       |
|                                   | KNO$_3$, (48, 98, 192 kg N ha$^{-1}$)                                                   |                      | no effect      |            |
| Deciduous forest soil, Austria    | KNO$_3$, (0, 10, 100, and 500 mg N kg$^{-1}$ soil)                                      | Ambient air (1.8     | Inhibition     | [15]       |
|                                   | (NH$_4$)$_2$SO$_4$, (0, 10, 100, and 500 mg N kg$^{-1}$ soil)                           | ppm)                 |                 |            |
| Mixed deciduous forest, England   | (NH$_4$)$_2$SO$_4$ (5 and 50 mM)                                                        | Ambient air           | No effect      | [100]      |
|                                   | NH$_4$Cl (5 and 50 mM)                                                                 |                      | NH$_4$Cl: 82-84% |            |
| Coniferous forest England         | (NH$_4$)$_2$SO$_4$ (15, 30, 60 or 120 mM)                                              | 10 and 1,000 ppm     | No effect      | [46]       |
|                                   | KNO$_3$ (15, 30, 60 or 120 mM)                                                         | 10 and 1,000 ppm     | 70% reduction  |            |
|                                   |                                                                                       |                      | 30% reduction  |            |
|                                   |                                                                                       |                      | 70% reduction  |            |
Deciduous forest, UK

| NaNO₃ (varying con.) | Ambient air or higher reduction by the NO$_3$ | [93] |
|----------------------|---------------------------------------------|------|
| NaNO₂ (varying con.) | 50,000 ppm                                  |      |
| NH₄Cl (varying con.) |                                             |      |

Japan coniferous forest

| KNO₃ (200 mg N kg$^{-1}$ soil) | NaNO₂ (varying con.) | 2.4 and 12.6 ppm | 86.6% reduction | [47] |
| Urea (200 mg N kg$^{-1}$ soil) |                             |                  | 75% reduction    |      |

Deciduous forest soil, USA

| NH₄NO₃ (100 Kg ha$^{-1}$) | Ambient air | 40-60% reduction | [91] |
| NH₄Cl (60 kg N ha$^{-1}$) |             |                  |      |
| KNO₃ (60 kg N ha$^{-1}$)  |             |                  |      |
| KNO₂ (60 kg N ha$^{-1}$)  |             |                  |      |

Rice field soil, Italy

| NH₄Cl (60 kg N ha$^{-1}$) | 1000ppm | 250% increased | [80] |
| NH₄Cl (60 kg N ha$^{-1}$) | 10000ppm| 4% decreased   |      |
| KNO₃ (60 kg N ha$^{-1}$)  | 1000ppm | 300% increased |      |
| KNO₂ (60 kg N ha$^{-1}$)  | 10000ppm| 8% decreased   |      |

Deciduous temperate forest soil, Italy

| NH₄Cl (60 kg N ha$^{-1}$) | 1000ppm | 4% reduced     | [80] |
| NH₄Cl (60 kg N ha$^{-1}$) | 10000ppm| 14% increased  |      |
| KNO₃ (60 kg N ha$^{-1}$)  | 1000ppm | 18% reduced    |      |
| KNO₂ (60 kg N ha$^{-1}$)  | 10000ppm| 100% increased |      |

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