https://doi.org/10.5194/egusphere-2023-2033
Preprint. Discussion started: 12 September 2023

Parent material influences soil abiotic N_2O production from chemical oxidation of hydroxylamine

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https://doi.org/10.5194/egusphere-2023-2033
Preprint. Discussion started: 12 September 2023

Abstract. Hydroxylamine (NH_2OH) and nitrite (NO_2^-) represent pivotal nitrification intermediates that substantially govern soil abiotic N_2O production. Yet, the intricate factors influencing the abiotic formation of N_2O from chemical reactions involving NH_2OH and NO_2^- remain uncertain. This study was designed to reveal the impacts of land use type and parent material on soil abiotic N_2O production in response to NH_2OH and NO_2^- amendments. Our investigation revealed that land use type exerted no significant influence on abiotic production of N_2O with NH_2OH and NO_2^- addition. Nevertheless, the parent material exhibited a notable (P < 0.01) effect on N_2O production intrigued by NH_2OH addition. Specifically, a markedly higher abiotic N_2O production from NH_2OH was observed in soils developed from Quaternary red clay than those derived from granite. Subsequent analysis demonstrated that the soils originating from Quaternary red clay displayed significantly higher manganese (Mn) content in comparison to those originating from granite. This finding consistently aligns with the close correlation between the abiotic N_2O production via chemical oxidation of NH_2OH and Mn content of soil. Furthermore, the site preference (SP) values for N_2O arising from NH_2OH and NO_2^- addition were 25-30‰ and around 20‰, respectively, aligning with the expected ranges characterizing ammonia oxidation and chemodenitrification processes. Our findings provide valuable insight into the distinct influence of parent material on soil abiotic N_2O production via chemical oxidation of NH_2OH, contributing to a better understanding of the underlying mechanisms, and highlight the significance of soil factors in regulating abiotic N_2O production within soil ecosystems.

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1 Introduction

Nitrous oxide (N_2O) holds a pivotal position in the atmosphere, both as a long-lived greenhouse gas and a precursor to nitric oxide, contributing to the depletion of stratospheric ozone (IPCC, 2007). Agricultural soil ecosystems contributed to 52% of anthropogenic N_2O emissions due to the large input of nitrogen (N) fertilizers (Tian et al., 2020). Recent findings have revealed that N_2O production in soil is influenced by both biotic and abiotic processes (Heil et al., 2016; Liu et al., 2019, 2018; Wei et al., 2022). Notably, abiotic processes may contribute to 29.3-37.7% of global total N_2O production (Wei et al., 2022). Hydroxylamine (NH_2OH) and nitrite (NO_2^-) emerge as critical N intermediates that significantly contribute to abiotic N_2O production. However, the intricate factors influencing abiotic N_2O production from these two intermediates ~~were~~ still under investigation.

Hydroxylamine and NO_2^- are two crucial intermediates within N cycle (Heil et al., 2016). Hydroxylamine can be released into the extracellular environment by ammonia-oxidizing microorganisms (Liu et al., 2017), methane-oxidizing microorganisms (Versantvoort et al., 2020), heterotrophic nitrifiers (Stouthamer et al., 1997; Verstraete and Alexander, 1972) and dissimilatory nitrate reduction to ammonium ~~microorganisms~~ (Giblin et al., 2013; Hanson et al., 2013). Subsequently, NH_2OH undergoes oxidation mediated by soil metal oxidants, such as manganese oxide (MnO_2) and ferric ion (Fe^{3+}), leading to the formation of N_2O. Additionally, NH_2OH can undergo fixation by soil organic matter to form compounds known as oximes (Porter, 1969). Considerable accumulation of NO_2^- has been observed during nitrification in soils due to the

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uncoupling of ammonia and nitrite oxidation (Duan et al., 2020a; Tierling and Kuhlmann, 2018). Nitrite can also accumulate during the denitrification process in anaerobic conditions (Burns et al., 1996). Remarkably, this accumulated NO_2^- undergoes chemical reactions with ferrous iron and soil organic matter, giving rise to the intriguing phenomenon known as chemodenitrification (Nelson and Bremner, 1970, Wei et al., 2020). And various chemical properties of soil, such as pH, Fe concentration and organic matter content, have been demonstrated to exert significant influences on chemodenitrification (Chalk and Smith, 2020).

Generally, parent materials exert a significant impact on soil formation processes, leading to alteration in various soil physical and chemical properties, including soil pH and organic matter concentration (Miller and Donahue, 1990). Additionally, it has been documented that land use types and agricultural management practices, such as the application of inorganic N fertilizers and organic manure, have a marked influence on physical and chemical properties of soils (Zhang and Xu, 2005). For instance, the long-term utilization of mineral fertilizers in orchard soils is frequently associated with soil acidification (Guo et al., 2010; Yan et al., 2018). Consequently, it is reasonable to anticipate variations in abiotic N_2O production in soils derived from different parent materials and those under divergent land use types.

In our study, we conducted a laboratory incubation experiment to investigate the impacts of land use type and parent material on abiotic N_2O production through the addition of NH_2OH and NO_2^-. Gamma (γ) sterilization and site preference (SP) techniques were used to reveal the contribution of abiotic processes to N_2O production

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from different sources (Casciotti et al., 2010; Jung et al., 2014; Sutka et al., 2008, 2006). Previous research has revealed that distinct SP values for N_2O produced through various microbial and chemical processes. Specifically, SP values for N_2O molecule yielded by purely cultured ammonia-oxidizing archaea (AOA) have been documented in the range of 13.1-34‰ (Jung et al., 2014), while ammonia-oxidizing bacteria (AOB) exhibit SP values spanning from 14.2‰ to 38.2‰ (Casciotti et al., 2010). Additionally, Sutka et al. (2008) found that fungal denitrification had SP values ranging from 22.8‰ to 40‰. In contrast, bacterial denitrification processes presented a low SP range, falling in the scope of -5.9-0.7‰ (Haslun et al., 2018; Lewicka-Szczebak et al., 2014; Sutka et al., 2006; Toyoda et al., 2005; Yamazaki et al., 2014). Furthermore, SP values of N_2O produced through chemical reactions has been identified spanning a wide range of 16.8-35.6‰ (Buchwald et al., 2016; Grabb et al., 2017; Heil et al., 2015, 2014; Jones et al., 2015). Notably, Duan et al. (2020a) reported that SP values of N_2O produced from chemical reactions involving NH_2OH and NO_2^-, ranging from 27.4‰ and 36.5‰ in greenhouse vegetable soils. Nevertheless, the understanding of SP values stemming from abiotic N_2O production due to NH_2OH and NO_2^- addition in different soils remains limited. Therefore, in this study, we hypothesized that: 1) soil parent material and land use type could exert an influence on abiotic N_2O production from chemical reactions involving NH_2OH and NO_2^- due to the variations of soil physico-chemical properties; 2) SP values of N_2O molecule yielded from chemical oxidation of NH_2OH and NO_2^- chemodenitrification fall in the scope of abiotically-produced N_2O in previous studies.

2 Materials and methods

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2.1 Sample collection and chemical analysis

Fifteen soils from five sites in mainland China (Fig. 1): Changsha (CS, 113°19′E, 28°33′N), Tongcheng (TC, 113°46′E, 29°13′N), Taoyuan (TY, 111°31′E, 29°14′N), Dangyang (DY, 111°48′E, 30°41′N) and Xianning (XN, 114°22′E, 30°0′N) were collected for this study, representing a range of land use types and parent materials. At each site, soils were collected from the top 20-cm layer. After removing litters and plant roots, soils were placed into sealed bags and transferred to the laboratory of Huazhong Agricultural University. Soils were naturally dried in a well-ventilated room, then passed through a 2-mm sieve and stored at room temperature.

Basic soil physicochemical properties were analyzed before the incubation. Briefly, the metallic elements were determined with inductively coupled plasma optical emission spectrometry (Agilent 5110 ICP-OES, Agilent, USA). Total carbon (C) and N contents were analyzed with an elemental analyzer (Flash*Smart* 11206125, Thermo Scientific Inc., USA). Ammonium (NH_4^+) and nitrate (NO_3^-) were extracted with 1 M KCl (dry soil: solution = 1:10 w/w) and shaken for 24 h and determined with ion chromatography (Thermo Scientific, USA). Soil pH was measured with a pH probe after shaking soil with 1 M KCl (dry soil: solution = 1:10 w/w). Dissolved organic C (DOC) content was analyzed with acid-base titration method, while dissolved organic N (DON) content was calculated by total dissolved N (determined by ultraviolet spectrophotometer) minus dissolved inorganic N (NH_4^- and NO_3^-). The detailed information of soil chemical properties is shown in Table 1.

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2.2 Soil sterilization

Half of the soils were irradiated ~~under~~ 20 kGy (^{60}Co) at Huada Biological Technology, Guangzhou, China. After that, the soils were rewetted with sterilized deionized water to reach around 40% water-holding capacity (WHC), and then incubated for 72 h at 28 °C. The sterilized soils (1.0 g) were used to check irradiation efficiency by plating the soil slurries (soil : water = 1:1) on sterilized R2A medium, incubating for 72 h, 28 °C. Neither bacteria nor fungi was observed, demonstrating that the irradiation process was successful.

2.3 Amendment of NH_2OH and NO_2^- solutions

Sterilized and non-sterilized soils (0.5 g) were weighed into 12-ml gas chromatography (GC) vials (labco, England). Then, sterilized deionized water was added to bring the soil moisture to 40% WHC. Vials were then incubated for 72 h at room temperature (25 ± 1 °C). After that, NH_2OH and $NaNO_2$ solution were filtered through 0.22-μm filters and added to the vials to bring the soil to 60% soil WHC. Then, the vials were sealed tightly and incubated for 7 h at 25 °C. The added NH_2OH and NO_2^- concentration were 5 mg N kg^{-1} soil and 50 mg N kg^{-1} soil, respectively. The concentration of added NO_2^- mimicked the maximum level that can ~~achieve~~ after N fertilizer application; as for the concentration of NH_2OH added, 10-times smaller than amended NO_2^- ~~and~~ much higher than its natural content, the relatively high dose is beneficial for producing significant amount of N_2O and for further study of SP determination. At the end of incubation, 20 ml 99.9% N_2 was injected to the vials and then 20-ml headspace gas was collected from

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the vials. N_2O concentration in the headspace gas was analyzed by using an Agilent 7890A Gas Chromatograph.

Conversion rates of added NH_2OH and NO_2^- to N_2O were also calculated to predict the relative role of NH_2OH and NO_2^- in N_2O production, which were calculated by the following equations

$$[N_2O\text{-}N, NH_2OH] - [N_2O\text{-}N, control] / [NH_2OH\text{-}N] \quad (1)$$

$$[N_2O\text{-}N, NO_2^-] - [N_2O\text{-}N, control] / [NO_2^-\text{-}N] \quad (2)$$

where [N_2O-N, NH_2OH] is the concentration of N_2O-N derived from NH_2OH amendment; [N_2O-N, control] the concentration of N_2O-N derived from the control and [N_2O-N, NO_2^-] the concentration of N_2O-N derived from NO_2^- amendment; [NH_2OH-N] and [NO_2^--N] represent the concentration of added NH_2OH and NO_2^-, respectively.

2.4 Analysis of ^{15}N site preference of N_2O

To further explore the pathways of N_2O generation, the isotope experiment was performed. Soil (0.5 g) was weighed into a 12-ml labco vial followed by the addition of NH_2OH and NO_2^- solution, and then the vial was sealed tightly. The vials were incubated at 25 °C for 7 hours and 20 ml headspace gas was collected following the same steps mentioned above, to determine the ^{15}N site preference of N_2O by using an isotope ratio mass spectrometer (Toyoda and Yoshida, 1999). $^{15}N^{bulk}$ and $^{15}N^{\alpha}$ were monitored, $^{15}N^{\beta}$ could be calculated by the following equations

$$\Delta^{15}N^{\beta} = {}^{15}N^{bulk} - \delta^{15}N^{\alpha} \quad (3)$$

Site preference is defined as

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$$SP = \delta^{15}N^{\alpha} - \delta^{15}N^{\beta} \quad (4)$$

2.5 Statistical analysis

Analysis of variance (ANOVA) was performed for analyzing the differences in N_2O emissions among non-sterile and γ-irradiated samples. Tukey's post-hoc tests were utilized to identify significant differences at $P < 0.05$. The ANOVA also evaluated the percentage of total variance explained by land use types and parent materials. Spearman correlation analysis was performed to find relationship between N_2O emissions and possible variables. All the statistical analyses were conducted in R (version 4.0.3, R Foundation for Statistical Computing, Vienna, Austria).

3 Results

3.1 Effects of parent material and land use on soil physicochemical properties

Parent material had significant ($P < 0.05$) effects on several soil properties, including soil texture composition, DOC content, NO_3^- content, Mn and Fe contents (Table 2, Fig. 2). Soils developed from late Pleistocene sediment ~~have~~ the ~~largest amount of~~ silt and clay content, as well as NO_3^- and Fe contents among all the parent materials. Soils developed from granite showed a characteristic of the largest proportion of sand and the ~~least~~ Fe content among all the parent materials. The Mn content in soils from Quaternary red earth was significantly ($P < 0.05$) higher than that from granite and late Pleistocene sediment.

For land use type, the most affected properties were silt content, soil pH, TC, TN and

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NH_4^+ content (Table 2). Forest soil had the least TC and TN content, and lowest pH among all the land use types. Tea fields had the largest NH_4^+ content, and paddy soil showed the highest pH and the largest TC and TN among all the land use types.

3.2 Effects of parent material and land use type on N_2O production with NH_2OH and NO_2^- addition

Parent material had a significant ($P < 0.05$) influence on N_2O production from soils amended with NH_2OH (Fig. 3a, 4a). Soils developed from Quaternary red clay and late Pleistocene sediment have relatively higher N_2O production (0.77-4.76 mg N kg^{-1} soil and 1.97-2.11 mg N kg^{-1} soil, respectively) with NH_2OH addition than those developed from granite (0.16-1.07 mg N kg^{-1} soil). In the treatments with NO_2^- addition, N_2O production from different parent materials were in the range of 0.18-0.82 mg N kg^{-1} soil and showed insignificant difference (Fig. 3b, 4b). Land use type showed little influence on N_2O emissions among the 15 soils with NH_2OH and NO_2^- addition (Fig. 4). Average N_2O production from the five land use types were in an approximate range (0.98-2.28 mg N kg^{-1} soil and 0.39-0.50 mg N kg^{-1} soil, respectively) with NH_2OH and NO_2^- addition.

The conversion ratios of NH_2OH to N_2O in soils from Quaternary red clay (50.9%) was significantly larger than that from late Pleistocene sediment (27.7%), and granite (7.2%) (Table 3). Around 90% of NH_2OH to N_2O came from abiotic processes in soils from Quaternary red clay, while the ratios were around 69% for soils from late Pleistocene sediment and granite. The effect of land use types on the conversion ratios

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of NH_2OH to N_2O was not significant, with ratios ranging from 16.5% to 34.6%. The conversion ratios of NO_2^- to N_2O was much smaller than that of NH_2OH to N_2O, with ratios ranging from 0.93% to 11.6%. Neither parent material nor land use types affected the conversion ratios of NO_2^- to N_2O. Most (68.4-98%) of N_2O with NO_2^- addition came from abiotic processes.

3.3 Effects of soil factors on N_2O production with NH_2OH and NO_2^- addition

In non-sterile samples, N_2O production was positively ($P < 0.05$) correlated to soil Fe and Mn content with NH_2OH addition, whereas N_2O production was negatively ($P < 0.05$) correlated to soil Fe content and pH with NO_2^- addition (Fig. 5).

In γ-irradiated samples, N_2O production was positively ($P < 0.05$) correlated to soil Mn content with NH_2OH addition. However, soil Fe content did not affect N_2O production significantly in γ-irradiated samples with NO_2^- addition (Fig. 5). Conversely, N_2O production was negatively ($P < 0.05$) correlated to soil pH with NO_2^- addition, but correlated to soil DON positively ($P < 0.05$) (Fig. 5).

3.4 Site specific ^{15}N isotopic signatures of N_2O

Soils named DY1, CS1, TC3, TC5 were used for stable isotopic signature determination. The SP values of N_2O ranged from 26.8‰ to 28.4‰ with NH_2OH addition in non-sterile soils, which fell in the scope of SP values from nitrification (Fig. 6). The SP value of N_2O ranged from 18.2‰ to 21.6‰ with NO_2^- addition in non-sterile soils, which fell in the scope of SP values from chemodenitrification (Fig. 6). γ sterilization had no effect on the SP values of N_2O with NH_2OH addition, with SP value ranged

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from 24.5‰ to 28.9‰. Nevertheless, γ sterilization had significant (P < 0.05) effects on the SP values of N_2O ~~with~~ NO_2^- addition, with SP ~~value ranged~~ from 20‰ to 22.7‰. The $\delta^{18}O$-N_2O values ranged from 31.7‰ to 34‰ ~~with~~ NO_2^- addition in non-sterile soils, which ~~also fell in the scope~~ of values derived from denitrification. Nevertheless, the $\delta^{18}O$-N_2O ~~value~~ ranged from 5.7‰ to 15.6‰ ~~with~~ NH_2OH addition in non-sterile soils, which was much lower than the values observed ~~in~~ nitrification.

4 Discussion

Parent ~~materials~~ and land use ~~types~~ may have significant ~~influences~~ on soil basic physicochemical properties. During soil formation, changes occur in soil physical and chemical properties, such as soil pH and organic matter concentration (Alfaro et al., 2017; Angst et al., 2018). Moreover, the mineralogy of parent material can interact with soil management practices, including land use types and long-term fertilization, thereby influencing basic soil properties (Stone et al., 2015; Sun et al., 2016). In this study, parent materials had a significant influence on soil texture. Quaternary red clay and late Pleistocene sediment resulted in fine-textured soil while granite led to coarse-textured soil, which was similar to previous studies (Deng et al., 2015; Hu et al., 2003). In addition, significant differences in Mn and Fe contents across parent materials were observed. Soils developed from Quaternary red clay had the highest Mn content while soils ~~generated~~ from granite had the lowest Mn and Fe contents, which was consistent with earlier studies (Blume and Schwertmann, 1969; Bromfield et al., 1983). For total C and total N as well as NH_4^+-N content, land use types explained more variance than

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parent materials which was consistent with the study of Zhang et al. (2018). However, we observed significant difference in soil pH across land use types, which was not aligned with previous studies (Lauber et al., 2008; Zhang et al., 2018).

The conversion rates of NH_2OH to N_2O in the sterilized acidic soils ranged from 3.7% to 84.8% (data not shown), most of the results were fallen in the scope of earlier reports. Bremner et al. (1980) reported that NH_2OH addition increased N_2O production in sterilized soils, with conversion rates of NH_2OH to N_2O varied from 34% to 80%. Recently, Liu et al. (2019) revealed the conversion rates of NH_2OH to N_2O fell in the scope of 73-93%. Interestingly, we observed that parent material had significant effects on N_2O production from NH_2OH addition. Soils developed from Quaternary red clay had the highest N_2O production and late Pleistocene sediment the second largest, while granite led to the lowest N_2O yield. This was mainly attributed to the much higher Mn content in Quaternary red clay and high concentration of Fe in late Pleistocene sediment than granite. Furthermore, Spearman correlation coefficients suggested that the conversion rates of NH_2OH to N_2O were positively correlated to Mn and Fe content in soils (Fig. 5), which were consistent with the results shown by Bremner et al. (1980) and Liu et al. (2019).

Sterilization has affected the conversion rates of NH_2OH to N_2O significantly, with decrease of N_2O production after γ sterilization. Moreover, SP values were found in the range from 25‰ to 29‰ in both sterilized and unsterilized soils (Fig. 6), which were similar with the results of Jung et al. (2014) that SP values of N_2O produced by AOA ranged from 13.1‰ to 34‰. However, Heil et al. (2015) demonstrated SP values of

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34.1 ± 1.9‰ from a sterilized cropland soil with NH_2OH addition. In an earlier study, Heil et al, (2014) reported that the SP value of N_2O generated from chemical reactions with NH_2OH ranged from 33.9‰ to 35.6‰. Based on our findings, we inferred that N_2O produced from NH_2OH in acidic soils can be attributed to the combination of archaeal ammonia oxidation and abiotic reactions. However, further research is still necessary to fully elucidate factors influencing SP values in abiotic N_2O production pathways.

Regarding the conversion of NO_2^- to N_2O, NO_2^- addition increased N_2O production with conversion rates of 4.3% to 16.3%, which were in lined with the results of Venterea et al. (2003). Nevertheless, parent materials and land use types did not affect the conversion of NO_2^- to N_2O significantly. Correlation analysis showed that the conversion rates of NO_2^- to N_2O were negatively correlated to pH values and Mn content, and positively correlated to DON and C content (Fig. 5). Similar results were revealed by Liu et al. (2019) that soil properties, such as DOM, Fe, and Mn content, had strong effects on the contribution of NO_2^- to N_2O formation. Moreover, significant difference of N_2O production following NO_2^- addition was observed among non-sterile and sterilized acidic soils, which was probably due to the increased soil dissolved organic matter caused by γ sterilization (Berns et al., 2008). Nonetheless, SP values indicated that N_2O converted from NO_2^- was produced mainly via abiotic pathways. Our results were in line with the results reported by Liu et al. (2019), who found irradiation of soil had little influence on N_2O production in strongly acidic forest soils following NO_2^- addition.

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In supporting, SP values of N_2O produced from soils amended with NO_2^- were found in the range of 18‰ to 23‰ (Fig. 6), which were consistent with the results reported by Jones et al. (2015), who revealed that SP values of N_2O formed via NO_2^- reduction by Fe (II) were in the range of 10.0-22‰. Similarly, Wei et al. (2017) reported that N_2O generated from NO_2^- reduction with soil organic matter fractions at pH of 3.4 had a SP value of 20.27-26.14‰. Whereas, Toyoda et al. (2005) found that N_2O produced via NO_2^- reduction with trimethylamine-borane had a constant SP of approximately 30‰. Moreover, higher SP values of N_2O abiotically produced were reported by Heil et al. (2014), which were in the range of 34-35‰. It seems like SP values of N_2O chemically produced are in the relatively high-value scopes. However, $\delta^{18}O$-N_2O values measured in this study were lower than previously reported results, implying that a weaker fractionation during chemical reactions with NH_2OH in both non-sterile and sterilized soils. In conclusion, SP values obtained from this experiment shed some light in abiotic N_2O production processes with application of NO_2^- and NH_2OH to the same soil.

5 Conclusions

Our findings underscore the significant influence of parent materials on soil physicochemical properties, particularly Mn contents, and its potential effect on abiotic pathways to N_2O production in acidic soils. Furthermore, the accumulation of NH_2OH and NO_2^- amplifies the role of abiotic mechanisms, with their influence tightly linked to specific physicochemical properties of the soil. SP values reveal the contribution of abiotic processes of NH_2OH and NO_2^- to N_2O production in the tested soils additionally.

Further investigation should focus on the effects of parent materials on the activity and abundance of microbial community and portion of biotic N_2O production to total N_2O production.

Code availability. The R codes used for (bio)statistical analyses are available upon request.

Author contributions. Shurong Liu: Conceptualization, Methodology, Supervision, Writing-Review & Editing. Suyun Li: Investigation, Data curation, Writing-Original Draft, Writing-Review & Editing, Visualization. Cai Gan: Resources, Investigation, Writing-Original Draft. Danni Cai: Investigation. Jiani Ma: Writing-Review and Editing.

Competing interests. The contact author has declared that none of the authors has any competing interests.

Acknowledgements. This research was supported by Province natural science fund of Guangdong (2022A1515010786) to S. L. We sincerely appreciated the anonymous reviewers and editors for their critical and valuable comments to help improve this manuscript.

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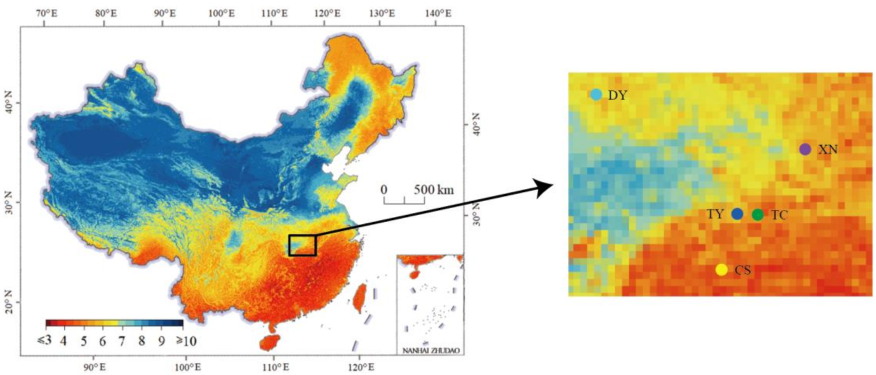

Figure 1. The geographical locations of the five sampling sites: Dangyang (DY), Xianning (XN), Taoyuan (TY), Tongcheng (TC) and Changsha (CS).

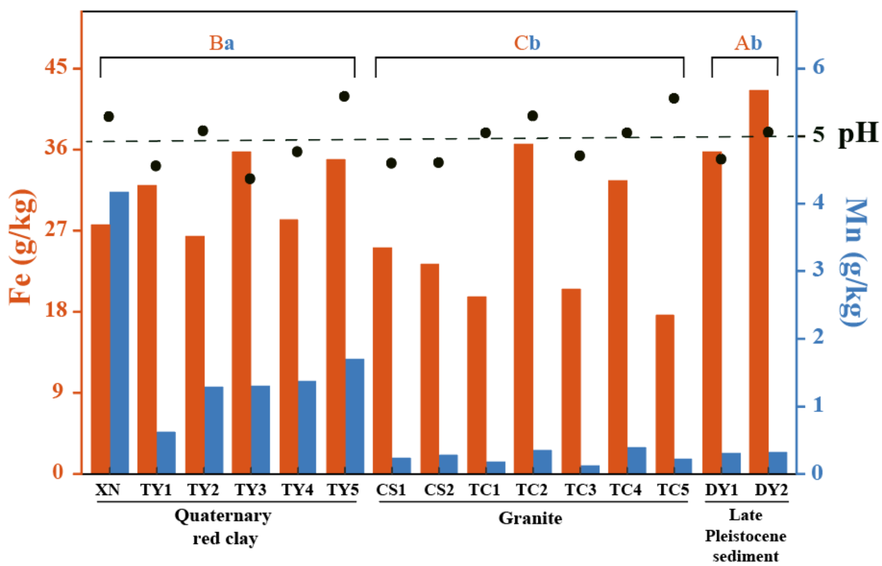

Figure 2. Manganese (Mn), iron (Fe) contents and pH values of the fifteen soils. Capital and lowercase letters indicate significant difference in Fe and Mn contents, respectively. The difference in pH values is insignificant.

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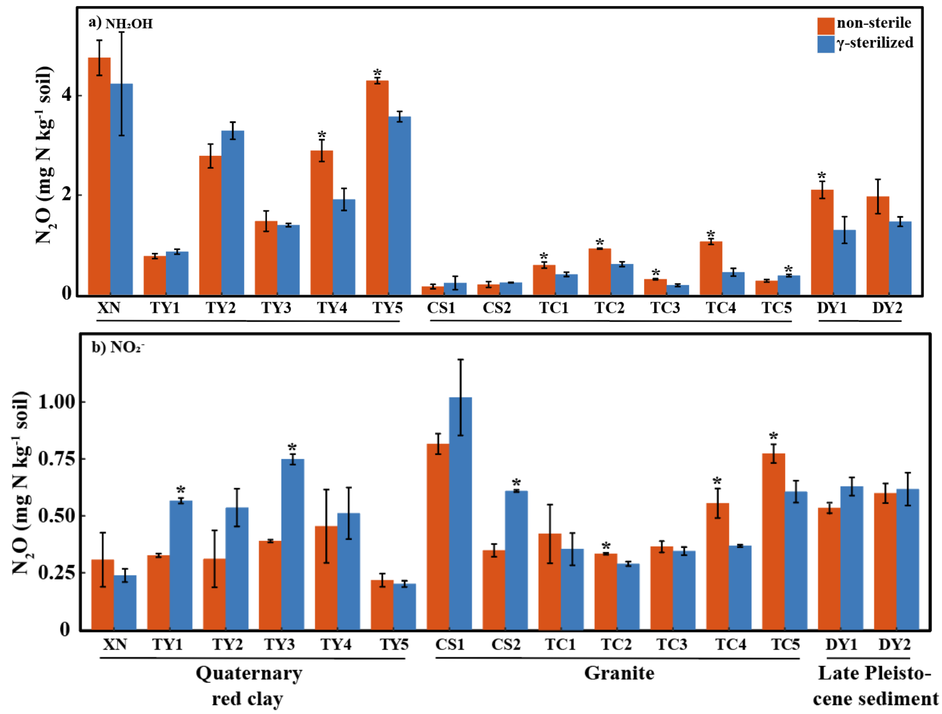

Figure 3. Net N_2O production in fifteen soils with and without γ sterilization after addition of NH_2OH (a) and NO_2^- (b) by parent material. Asterisks indicate significantly ($P < 0.05$) higher values between non-sterile and γ-sterilized samples at one soil. Error bars represent standard deviation (SD, n = 3).

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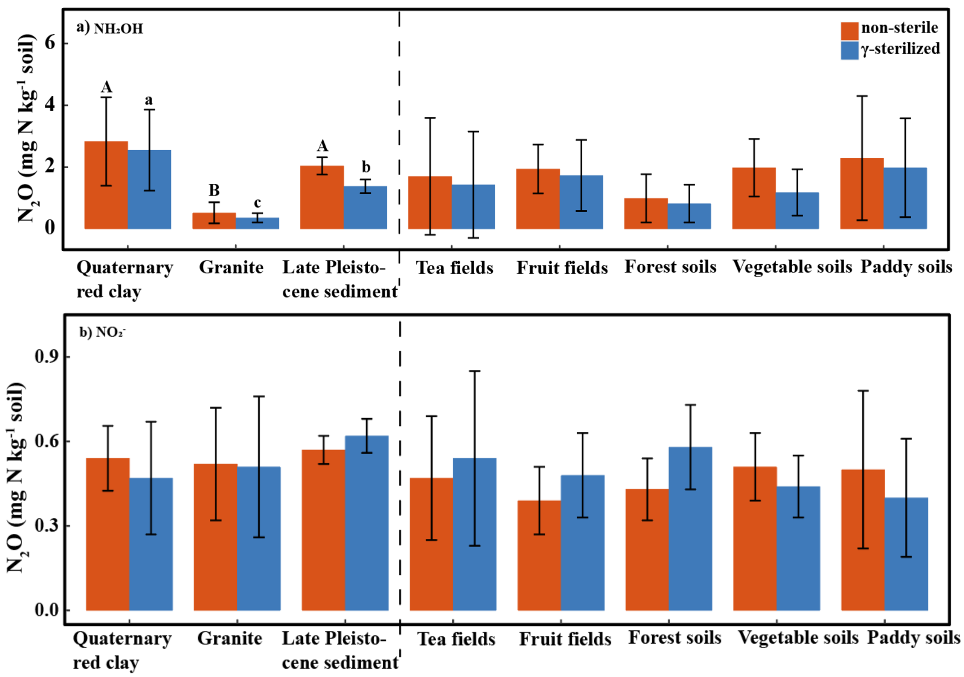

Figure 4. Emissions of N_2O from soils amended with NH_2OH (a) and NO_2^- (b), which were sorted by parent material and land use type. Capital and lowercase letters indicate significant difference among non-sterile and γ-sterilized soils, respectively. No letters indicate insignificant difference.

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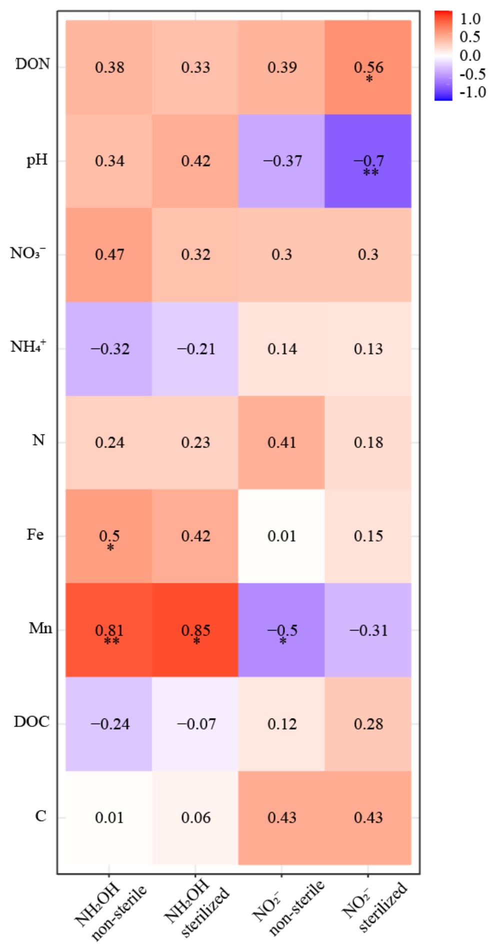

Figure 5. Spearman correlation coefficients between soil N_2O production from different amendments and soil properties after 7-hour incubation. One asterisk represents a significance under $P < 0.05$; two asterisks represent a significance under $P < 0.01$.

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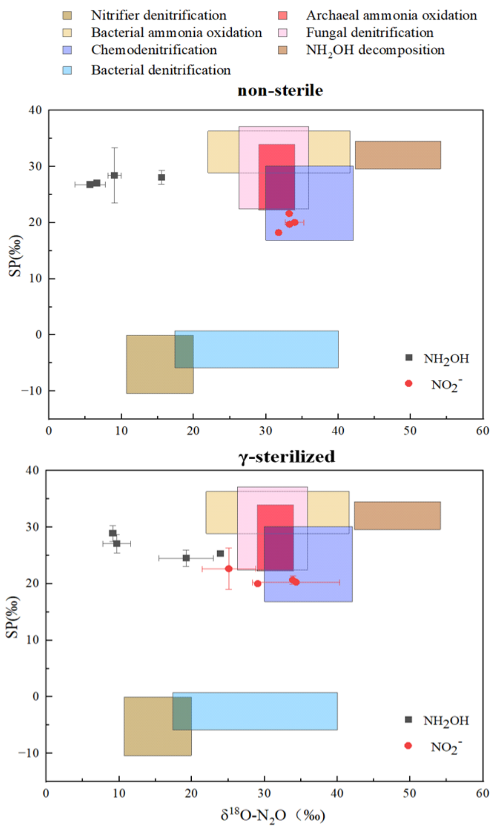

Figure 6. Relationships between the SP and δ^{18}O values of N_2O in non-sterile and γ-sterilized soils after NH_2OH and NO_2^- addition. The selected soils are DY1, CS1, TC3, TC5. Parameter references are listed as follow: (Buchwald et al., 2016; Frame and Casciotti, 2010; Grabb et al., 2017b; Haslun et al., 2018; Heil et al., 2015, 2014; Jinuntuya-Nortman et al., 2008; Jones et al., 2013; Jung et al., 2014; Lewicka-Szczebak et al., 2014; Maeda et al., 2015; Ostrom et al., 2010; Rohe et al., 2017; Santoro et al., 2011; Sutka et al., 2008, 2006; Toyoda et al., 2005; Well and Flessa, 2009; Yamamoto, 2017; Yamazaki et al., 2014; Yang et al., 2014).

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Table 1. Summary of soil properties in this study.

Parent material	Land use	Soils	Silt (%)	Clay (%)	Sand (%)	pH	TC (g/kg)	TN (g/kg)	DOC (mg/kg)	DON (mg/kg)	NH_4^+ (mg/kg)	NO_3^- (mg/kg)
Quaternary red clay	Tea	XN	9.9	33.13	56.97	5.29	9.64	1.043	13.43	9.91	3.41	9.17
	Tea	TY1	37.08	34.47	28.45	4.56	12.31	0.931	17.92	7.07	8.33	3.61
	Fruit	TY2	52.51	31.88	15.61	5.08	8.77	0.97	14.86	9.08	5.88	5.88
	Forest	TY3	40.43	40.87	18.7	4.37	11.13	0.97	16.62	11.59	18.52	6.89
	Vegetable	TY4	51.87	33.9	14.23	4.77	8.96	1.022	15.35	7.64	5.71	7.65
	Paddy	TY5	43.48	35.33	21.2	5.59	8.2	1.022	13.24	7.64	18.85	5.99
Late Pleistocene sediment	Tea	CS1	35.54	31.7	32.76	4.6	12.24	1.204	24.38	40.98	89.29	13.83
	Forest	CS2	21.32	26.96	51.73	4.61	3.26	0.255	13.28	5.05	4.06	3.94
	Tea	TC1	23.22	26.47	50.32	5.05	4.58	0.413	10.99	3.92	9.2	2.56
	Fruit	TC2	4.59	19.19	76.22	5.3	3.45	0.399	16.9	5.12	8.72	3.63
	Forest	TC3	30.64	22.54	46.81	4.71	1.25	0.207	12.15	3.06	3.9	5
	Vegetable	TC4	35.13	33.5	31.37	5.05	11.74	1.166	13.51	8.33	5.66	8.66
	Paddy	TC5	6.85	29.92	63.23	5.56	20.78	1.876	22.97	6.27	14.94	3.07
Granite	Fruit	DY1	48.49	39.71	11.79	4.66	8.91	0.999	12.86	24.3	1.98	20.19
	Forest	DY2	43.23	51.52	5.25	5.06	8.27	0.939	12.67	14.36	3.54	15

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Table 2. One-way ANOVA ($P < 0.05$) of soil physicochemical properties including dissolved organic carbon (DOC), total carbon (TC), total nitrogen (TN), total ferric ion (Fe), total manganese (Mn), silt, clay and sand content, etc..

Variables	Silt (%)	Clay (%)	Sand (%)	pH	TC (g/kg)	TN (g/kg)	DOC (mg/kg)	DON (mg/kg)	NH_4^+ (mg/kg)	NO_3^- (mg/kg)	Fe (g/kg)	Mn (g/kg)
Quaternary red clay	37.59a	35.18b	27.23b	4.94a	9.87a	0.99a	15.24a	8.82b	10.12a	6.53b	30.80b	1.74a
Granite	19.45b	27.18c	53.37a	5.04a	8.13a	0.69a	16.31a	10.39b	19.39a	5.81b	21.34c	0.25b
Late Pleistocene sediment	45.86a	45.62a	8.52c	4.86a	8.59a	0.97a	12.77a	19.33a	2.76a	17.60a	39.80a	0.26b
Tea field	22.78b	31.13a	46.10a	4.88bc	9.68b	0.90bc	16.68a	15.47a	27.56a	7.29a	26.08a	1.30a
Fruit field	33.03ab	30.06a	36.91a	5.01b	7.31a	0.82bc	14.88a	12.84a	5.53b	9.90a	25.05a	0.61a
Forest soil	34.84ab	37.50a	27.65a	4.69c	6.42b	0.64c	13.68a	7.94a	8.04ab	7.43a	29.43a	0.52a
Vegetable soil	41.82a	33.66a	24.52a	5.13b	10.07b	1.09ab	14.43a	8.96a	5.10b	8.53a	29.51a	0.80a
Paddy soil	25.16ab	32.62a	42.21a	5.58a	14.49a	1.45a	18.10a	6.95a	16.90ab	4.53a	26.29a	0.95a

556 **Table 3.** The conversion ratios (%) of NH_2OH and NO_2^- to N_2O and the contribution ratios (%)
557 of abiotic N_2O production to the total N_2O emission with NH_2OH and NO_2^- addition.

	MH_2OH		**NO_2^-**	
	Conversion ratio(%)	**Abiotic contribution (%)**	**Conversion ratio(%)**	**Abiotic contribution (%)**
Quaternary red clay	50.9 ± 26.1a	90.0 ± 16.4a	9.3 ± 4.0a	91.2 ± 21.2a
Granite	7.2 ± 3.1c	69.3 ± 25.7b	10.3 ± 5.0a	87.5 ± 15.2a
Late Pleistocene sediment	27.7 ± 4.4b	69.2 ± 15.4b	12.5 ± 1.2a	98.2 ± 4.1a
Tea field	28.9 ± 34.3a	76.6 ± 28.4ab	10.9 ± 6.2a	98.0 ± 27.1a
Fruit field	34.6 ± 23.0a	77.4 ± 20.8ab	9.7 ± 3.1a	95.6 ± 6.5a
Forest soil	16.5 ± 12.2a	83.4 ± 19.5a	11.6 ± 3.0a	97.9 ± 3.7a
Vegetable soil	23.7 ±15.0a	54.1 ± 14.1b	8.8 ± 2.1a	68.4 ± 28.4b
Paddy soil	39.7 ± 32.0a	91.6 ± 8.6a	8.1 ± 4.1a	85.9 ± 9.6ab

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