

Article

Nitrous Oxide Emission and Crop Yield in Arable Soil Amended with Bottom Ash

Do-Yeong Hur¹, Sung-Un Kim^{1,2}, Hyeon-Cheol Park¹, Keun-Ki Kim¹, Hong-Ju Son¹ , Kwang-Min Lee¹ , Yu-Jin Kim¹  and Chang-Oh Hong^{1,2,*}

¹ Department of Life Science and Environmental Biochemistry, Pusan National University, Miryang 50463, Korea; hdy2953@pusan.ac.kr (D.-Y.H.); cross9279@pusan.ac.kr (S.-U.K.); hcpark@pusan.ac.kr (H.-C.P.); kkkim@pusan.ac.kr (K.-K.K.); shjoo@pusan.ac.kr (H.-J.S.); leekm@pusan.ac.kr (K.-M.L.); yjkim2020@pusan.ac.kr (Y.-J.K.)

² Life and Industry Convergence Research Institute, Pusan National University, Miryang 50463, Korea

* Correspondence: soilchem@pusan.ac.kr; Tel.: +82-55-350-5548

Abstract: Bottom ash (BA), a byproduct of coal combustion from electric power plants with a porous surface texture and high pH, may influence the physical and chemical properties of upland arable soil associated with nitrous oxide (N₂O) emission from upland soil. This study evaluated the use of BA in mitigating N₂O emissions from upland arable soil and increasing the crop yield. In a field experiment, N₂O emitted from the soil was monitored weekly in a closed chamber over a 2-year period (2018–2019). BA was applied to upland soil at the rates of 0, 200, and 400 Mg·ha⁻¹. Cumulative N₂O emission significantly decreased with increasing BA application rate; it decreased by 55% with a BA application rate of 400 Mg·ha⁻¹ compared with the control. Yield-scaled N₂O emission decreased with increasing BA application rates of up to 200 Mg·ha⁻¹. Water-filled pore spaces (WFPS) were 70.2%, 52.9%, and 45.3% at the rates of 0, 200, and 400 Mg·ha⁻¹, respectively, during the growing season. For economic viability and environmental conservation, we suggest that BA application at a rate of 200 Mg·ha⁻¹ reduces N₂O emissions per unit of crop production.

Keywords: bottom ash; nitrous oxide; yield-scaled nitrous oxide emission



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1. Introduction

Nitrous oxide (N₂O) has 298-fold higher global warming potential than carbon dioxide (CO₂) over a 100-year time horizon, contributing up to 6% of global warming [1,2]. Agricultural soils are the primary source of global anthropogenic N₂O emissions, accounting for approximately 60% of the total N₂O emissions [3], primarily owing to the application of synthetic nitrogen (N) fertilizers and manures.

N₂O is mainly produced as a result of microbial nitrification and denitrification in soils. Although nitrification requires O₂, denitrification relies on its absence or limitation and is attributed to anoxic conditions. Water-filled pore spaces (WFPSs) of soils can be an indicator of the activity of aerobic and anaerobic microorganisms. Here, higher WFPS values indicate that more air replaced by water in pores, thus removing O₂ from the soil. Changes in WFPS value comprise an important factor influencing N₂O emissions from arable soils [4–7]. Nitrification that requires NH₄⁺ as an inorganic N substrate for aerobic respiration is the predominant process of N₂O production from soils with a WFPS value of <60%. Conversely, denitrification that requires NO₃⁻ as an alternative electron acceptor for anaerobic respiration is a predominant process of N₂O production from soils with a WFPS value of >60% [6,8–12]. N₂O emissions increase dramatically when both processes occur simultaneously as the soil WFPS value increases to almost 60% [13]. Soil pH is considered a critical factor for controlling various chemical and biological soil properties. Furthermore, soil pH has a significant effect on microbial processes responsible for the production and consumption of N₂O in soils [14]. Primarily, the last step of denitrification (N₂O → N₂)

is strongly controlled by soil pH. The reduction of N₂O to N₂ linearly increases with increasing soil pH [15,16]. In addition, adjustment of the soil pH increases the abundance of N₂O reductase genes (NosZ) at high pH values and decreases soil N₂O emissions [17].

Coal ash is a byproduct of coal combustion in electric power plants. In 2020, more than 10 million tons of coal ash were generated globally, which are associated with storage, processing, and reclamation costs as well as near saturation of coal-fired power plants [18]. Coal ash is categorized into fly ash (FA) and bottom ash (BA) based on particle size. The recycling rate of FA, a concrete additive and raw cement material or mixture, is 64% of the total FA recycle amount. Of the total BA, 37% is recycled as a concrete additive, and the remaining 60% is used as landfill in the Republic of Korea [19].

BA is larger and more irregular than FA and contains pores and cavities [20], has alkalinity close to pH 9, and primarily comprises silicate (SiO₂), aluminum oxide (Al₂O₃), and iron oxide (Fe₂O₃) [21,22]. Albuquerque [23] reported that BA has highly available nutrients that can be applied to the soils to reverse nutrient depletion, and BA properties improve soil quality by increasing soil aeration [23]. Several studies have reported that using BA for soil amendment improves crop productivity without heavy metal toxicity [24–26].

The application of BA, which is porous and alkaline and adds plant nutrients to the arable soil, may change the physical and chemical properties of the soil, such as WFPS value, soil pH, and nutrient supply that influence the N₂O emission and crop yield. Based on previous studies [7,13,15–17,23–26], this study presents three hypotheses: (1) the application of porous BA decreases the bulk density and WFPS value of soil to make soil conditions unfavorable for microorganisms associated with N₂O-production processes, such as nitrification and denitrification; (2) applying alkaline BA increases soil pH, promotes reduction of N₂O to N₂, and decreases N₂O emission; and (3) BA containing various elements provides available nutrients for plant growth and increases crop yield. Therefore, this study was conducted to evaluate the effects of BA application on mitigating N₂O emission and increasing crop yield in upland soil within a radish–maize cropping system for 2 consecutive years.

2. Materials and Methods

2.1. Experimental Site

A field experiment was performed on arable soil located in Cheonghak-ri, Samrangjin-eup, Miryang, Gyeongnam Province, South Korea (35°26′59.9″ N, 128°48′29.6″ E). Single cropping of paddy rice cultivation was performed in the study site for approximately 10 consecutive years before the experiment. The experimental field was drained on 1 September 2017, for harvesting rice, which then remained drained throughout the experimental period. The study was conducted over 2 years: from April 2018 to March 2019 and from April 2019 to March 2020 for Years 1 and 2, respectively. Rotational cultivation of radish (*Raphanus sativa* L.) and corn (*Zea mays* L.) was performed in upland soil.

The average mean temperature and rainfall for the experimental period were 14.1 °C and 1259 mm, respectively. The soil belongs to the Chilgok series (fine loamy, mixed, mesic family of Anthraquic Hapludalfs) and is well drained with a 2% slope. The specific physical and chemical properties of the study soil are summarized in Table 1.

Table 1. Chemical and physical properties of studied soil ($n = 3$).

Ph (1:5 H ₂ O)	Electrical Conductivity (ds·m ⁻¹)	Bulk Density (g·cm ⁻³)	Organic Matter (g·kg ⁻¹)	Total Nitrogen (g·kg ⁻¹)	Available Phosphate (mg·kg ⁻¹)	NH ₄ (mg·kg ⁻¹)	NO ₃ (mg·kg ⁻¹)	Soil Texture
5.47 ± 0.25	0.35 ± 0.15	1.26 ± 0.12	16.4 ± 0.18	1.05 ± 0.15	215 ± 20	5.1 ± 0.14	2.9 ± 0.12	Silt loam

$n = 3$: three replicates.

2.2. Characterization of BA

The BA used in this study was a byproduct of the coal-fired power plant from Korea South-East Power Co., Ltd. in Yeongheung, Republic of Korea. The BA was produced at a high temperature of approximately 1200 °C. Expanded X-ray diffraction (XRD) pattern analysis was performed to characterize the BA crystal structures as shown in Figure 1a. The surface morphology of BA and its physical and chemical properties are shown in Figure 1b and Table 2, respectively. BA had a pH of 9.3, total carbon of 6.6 g·kg⁻¹ and total nitrogen of 0.9 g·kg⁻¹. It was porous (58% porosity) and had a large surface area (10.1 m²·g⁻¹).

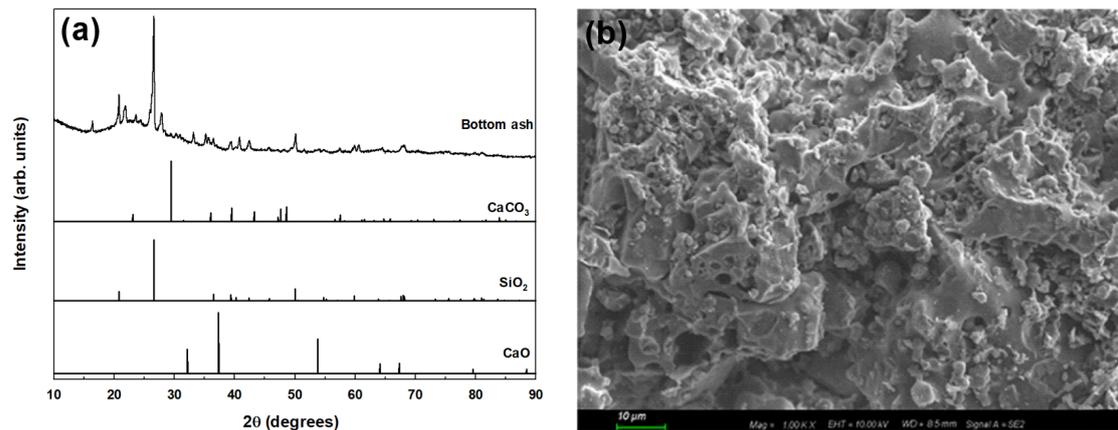


Figure 1. Expanded X-ray diffraction patterns (a) and surface morphology of bottom ash analyzed by SEM-EDS (b) used in this study.

Table 2. Chemical and physical properties of bottom ash used in this study ($n = 3$).

pH (1:5, KCl)	Electrical Conductivity (dS·m ⁻¹)	Total Carbon (g·kg ⁻¹)	Total Nitrogen (g·kg ⁻¹)	Total Phosphorus (mg·kg ⁻¹)	SiO ₂ (%)	CaCO ₃ (%)	CaO (%)	Surface Area (m ² ·g ⁻¹)	Porosity (%)
9.3 ± 0.19	1.4 ± 0.09	6.6 ± 0.4	0.9 ± 0.06	130 ± 17	60 ± 1.4	39 ± 0.9	1 ± 0.06	10.1 ± 0.12	58 ± 3.4

$n = 3$: three replicates.

Silicate (SiO₂) and calcium carbonate (CaCO₃) showed distinct characteristic peaks, indicating high purity and crystallinity. However, calcium oxide (CaO) showed several weak characteristic peaks. As shown during XRD pattern analysis, the relative percentages of SiO₂, CaCO₃, and CaO were 60%, 39%, and 1%, respectively.

2.3. Experimental Design and Crop Management

The BA was applied at the rates of 0, 200, and 400 Mg·ha⁻¹ to the experimental plots before crop cultivation on 1 April 2018. In total, 12 plots (3 m × 4 m for each plot) were arranged in a randomized complete block design with four replicates. First, radish seeds were sown and then maize seeds. Radish seeds were sown and harvested on 30 March and 29 May 2018, and 1 April and 21 May 2019, respectively. Maize was planted and harvested on 29 May and 18 September 2018, and 22 May and 28 August 2019. Urea, fused phosphate, and potassium chloride were applied at rates of N–P₂O₅–K₂O = 42–21–40 kg and 93–35–74 kg ha⁻¹ for radish and maize at base fertilizer application, respectively. Additional fertilizers such as urea and potassium chloride were applied at the rates of 74–0–40 and 93–0–0 kg ha⁻¹ for radish and maize, respectively. Base fertilizer application for radish and maize was conducted on 30 March and 29 May 2018, for Year 1 and on 1 April and 22 May 2019, for Year 2, respectively. Additional fertilizers for radish and maize were top-dressed on 1 May and 1 August 2018, for Year 1 and 13 May and 22 June 2019, for Year 2, respectively. BA and chemical fertilizers were spread on the soil surface and mixed into the soil using a rototiller to a depth of approximately 20 cm.

2.4. N₂O Emission Measurements

A closed chamber method [27] was used to measure N₂O emission from soil. Static PVC column chambers (headspace; 10.8 L, 25 cm diameter × 22 cm height) were placed at the center of each plot on 18 March 2018. The collar was placed between crops (radish and maize), and crops were not planted inside the chamber. All weeds that grew inside the chamber were removed during the experimental period. After 2 weeks of acclimation, the collar was closed with a fitted lid and air vent and rubber septa to collect gas samples. The plants grown inside the chamber were removed during the experimental period. Gas samples were collected using 30-mL syringes at 0, 20, and 40 min after covering the lid between 10:00 and 12:00 h throughout the year-long study period. Gas samples were collected once a week during the growing season of radish and maize, twice a week at special events, such as chemical fertilizer application and heavy rainfall, and once every 2 weeks during the fallow season. Gas sampling was performed throughout the study period from April 2018 to March 2020. The temperature in the chamber during gas sampling was measured using a portable thermometer (WT-1, Elitech, London, UK). The concentration of N₂O was analyzed using a gas chromatograph–mass spectrophotometer (GC-MS QP2020, Shimadzu, Kyoto, Japan). N₂O fluxes were calculated based on the slope of the linear increase in concentration during the chamber-closure period.

The N₂O flux was calculated using the following formula:

$$\text{N}_2\text{O flux (g}\cdot\text{ha}^{-1}\cdot\text{day}^{-1}) = \rho \times (V/A) \times (\Delta c/\Delta t) \times (273/T + 273) \times k \times a \quad (1)$$

where ρ is the gas density of N₂O (g·m⁻³), V is the volume of the chamber (m³), A is the soil area covered by the chamber (m²), $\Delta c/\Delta t$ is the rate of change in gas concentration in the chamber (g·m⁻³·min⁻¹), T is the average temperature in the chamber (°C), k is the time conversion coefficient (min·day⁻¹), and a (10,000 m²·ha⁻¹) is the area conversion coefficient.

Cumulative N₂O emission was calculated as follows (kg·N₂O·ha⁻¹):

$$\text{Cumulative N}_2\text{O emission (kg}\cdot\text{ha}^{-1}) = \sum \left(\frac{F_i + F_{i+1}}{2} \right) \times (t_{i+1} - t_i) \quad (2)$$

where F represents the N₂O flux (g·N₂O·ha⁻¹·day⁻¹), i is the i th measurement, and $(t_{i+1} - t_i)$ is the number of days between two adjacent measurements.

Yield-scaled N₂O emission was calculated as follows [28]:

$$\text{Yield-scaled N}_2\text{O emission (kg}\cdot\text{N}_2\text{O}\cdot\text{Mg)} = \text{cumulative N}_2\text{O emission (kg}\cdot\text{ha}^{-1})/\text{crop yield (Mg}\cdot\text{ha}^{-1}). \quad (3)$$

2.5. Soil Sampling and Analysis

In each plot, soil sampling was performed before starting the experiment, after base and additional fertilizer applications, and after crop harvesting in the radish and maize growing season. Samples of the soil layers at 0–15 cm depth were collected using a hand auger (5 cm diameter) and a core sampler (100 cm³) to measure soil bulk density. Samples were then air-dried, passed through a 2 mm sieve, and kept in a plastic zipper bag for chemical analysis. Soil pH and EC were measured in a 1:5 soil:distilled water suspension using a pH and electrical conductivity meter (Orion Star A215, Thermo Scientific Orion, Mansfield, TX, USA). Organic matter content was analyzed using the Walkley and Black method [29], total nitrogen was analyzed using the Kjeldahl method, and available phosphorus was analyzed using the Lancaster method (5 g soil was extracted with 20 mL of 0.33 M CH₃COOH, 0.15 M lactic acid, 0.03 M NH₄F, 0.05 M (NH₄)₂SO₄, and 0.2 M NaOH at a pH of 4.25) [30]. Analysis of soil inorganic nitrogen content was performed by adding 25 mL of 2 M KCl to 5 g of dry soil samples and shaking for 30 min, after which sample solutions were filtered using Whatman No. 2 filter paper. Ammonium nitrogen (NH₄⁺) was measured using the indophenol-blue colorimetric method [31], and nitrate-nitrogen (NO₃⁻) was measured using the brucine method [32].

Soil temperature and moisture at 10 cm soil depth were measured for each treatment at 3 h intervals using sensors (5TE Water Content, Temperature, and Electrical Conductivity, Decagon Devices, Inc., Pullman, WA, USA). Weather data, such as daily average temperature and precipitation during the experimental period were obtained from the local weather observation data (Aanderaa, Automatic weather station, xylem, Bergen, Norway) of the Korean Meteorological Administration (latitude: 35°29' N; longitude: 128°44' E; distance from the experimental field: 7.48 km).

WFPS (%) was calculated every day for a year using the following equation:

$$\text{WFPS (\%)} = (\theta / \text{soil porosity}) \times 100 \quad (4)$$

where θ is the volumetric moisture content ($\text{m}^3 \cdot \text{m}^{-3}$). Soil porosity ($\text{m}^3 \cdot \text{m}^{-3}$) was calculated using a particle density value of $2.65 \text{ g} \cdot \text{m}^{-3}$ and soil bulk density values. Bulk density was determined using soil samples of all plots collected from a depth of 0–15 cm once a month during the study period. Samples were collected by a fixed-volume core (94.64 cm^3) and dried at $105 \text{ }^\circ\text{C}$.

2.6. Statistical Analysis

Statistical analysis was performed using R software package V. 3.6.2 (<http://www.R-project.org>, accessed on 12 June 2020). The mean values of cumulative N_2O emissions, radish and maize yields, and yield-scaled N_2O emissions were compared using pairwise comparison. Differences among parameters were determined using two-way analysis of variance. Application rate (R) and year (Y) were considered to be fixed effects. Owing to application rate \times year interactions for cumulative N_2O emission during radish and maize growing seasons, each year was separately analyzed. The least significant difference was used for multiple comparisons between the means and performed only when the F-test result was significant ($p < 0.05$).

3. Results

3.1. N_2O Flux

N_2O flux patterns differed with air temperature patterns over the study period, as shown in Figure 2a,b. Although flux was relatively low during the cold and dry fallow season, it did not peak as air temperature reached its maximum in the August of both Years 1 and 2. N_2O flux peaks appeared only during the radish and maize growing seasons in Years 1 and 2 when basal and additional N fertilizers were applied, but peaks did not appear during fallow seasons. More N_2O flux peaks appeared during the maize growing season than during the radish growing season in Years 1 and 2.

Daily WFPS values varied over 2 years and increased after high rainfall and irrigation events during this period (Figure 2c). BA application rate resulted in a change in soil WFPS values. Daily WFPS values of soil amended with $0 \text{ Mg} \cdot \text{ha}^{-1}$ of BA were always the highest, followed by those of soil amended with 200 and $400 \text{ Mg} \cdot \text{ha}^{-1}$ of BA over the 2 years.

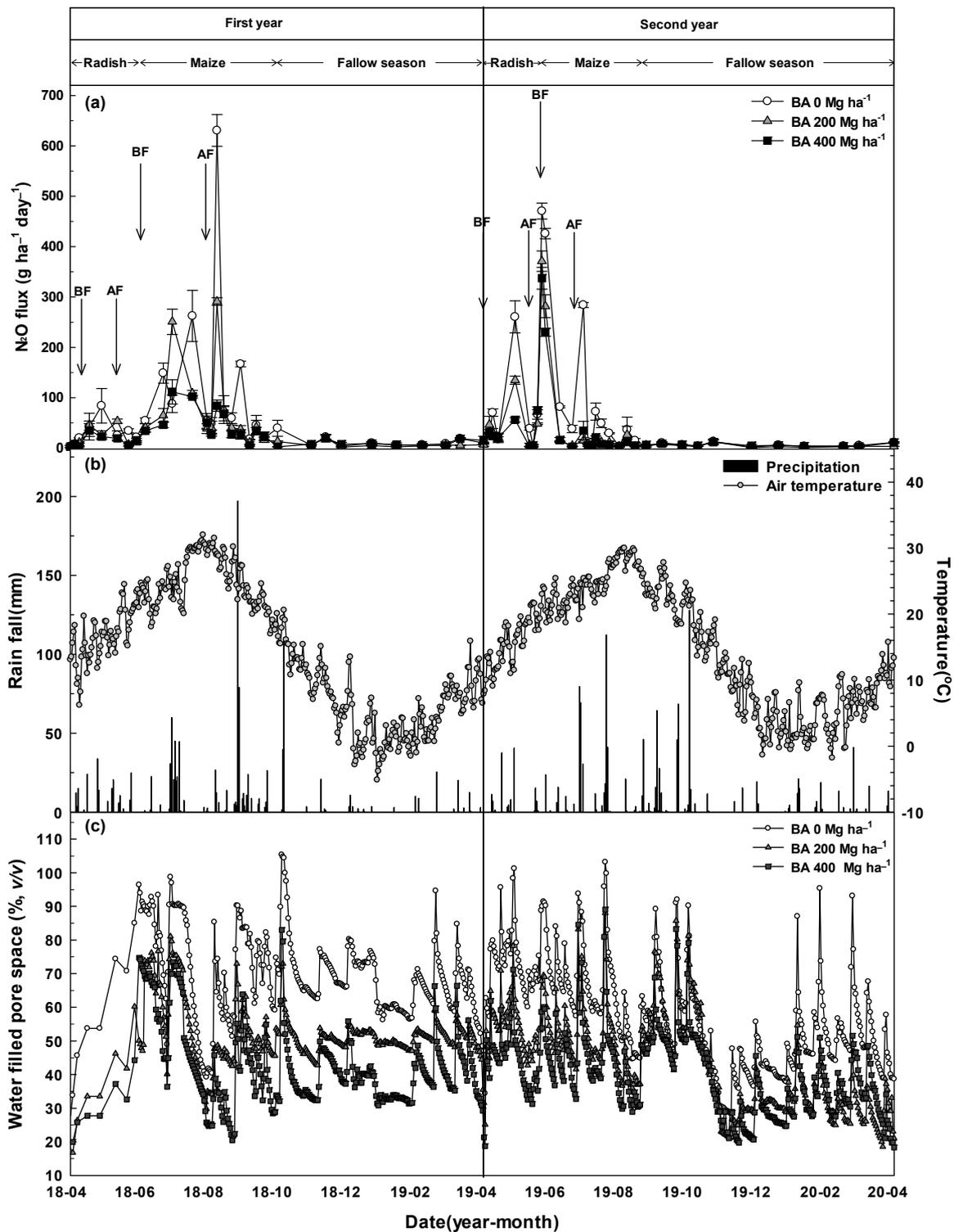


Figure 2. Nitrous oxide flux (a), daily air temperature, precipitation (b), and water-filled pore space (c) after the application of bottom ash at various rates for 2 years. Arrows in the graph represent fertilizer addition and irrigation, and BF and AF denote base and additional fertilizer applications, respectively.

3.2. Cumulative N_2O Emission

There was a significant $R \times Y$ interaction for cumulative N_2O emission during the radish growing season (Table 3). Cumulative N_2O emission during the radish growing season decreased significantly following application of $400\ Mg\cdot ha^{-1}$ of BA in Year 1 and with increasing BA application rate in Year 2 (Table 4).

Table 3. Analysis of variance and probability value of cumulative nitrous oxide (N₂O) emissions, chemical and physical properties of soil, and crop yields.

Source of Variation	Application Rate (R)	Year (Y)	R × Y
Df	2	1	2
Cumulative N ₂ O emission during radish growing season	<0.001	<0.001	0.0232
Cumulative N ₂ O emission during maize growing season	<0.001	<0.001	0.0166
Cumulative N ₂ O emission during entire year	<0.001	NS	NS
Averaged bulk density	<0.001	<0.001	NS
Averaged water-filled pore space during radish growing season	<0.001	<0.001	<0.001
Averaged water-filled pore space during maize growing season	<0.001	NS	NS
Averaged water-filled pore space during entire year	<0.001	NS	NS
Soil pH	<0.001	NS	NS
Electrical conductivity	NS	NS	NS
Organic matter	NS	<0.001	NS
Total nitrogen	NS	<0.001	NS
Av. P ₂ O ₅	NS	<0.001	NS
Exchangeable K	NS	NS	NS
Exchangeable Ca	NS	NS	NS
Exchangeable Mg	NS	NS	NS
Bulk density	<0.001	<0.001	NS
Ammonium (NH ₄ ⁺) in soil	0.0224	<0.001	NS
Nitrate (NO ₃ ⁻) in soil	<0.001	NS	NS
Radish yield	NS	<0.001	NS
Maize yield *	NS	NS	NS
Radish yield-scaled N ₂ O emission	<0.001	<0.001	NS
Maize yield-scaled N ₂ O emission	0.0015	NS	NS
Total yield-scaled N ₂ O emission	<0.001	NS	NS

Maize yield *: grain yield of maize. NS: not significant.

Table 4. Cumulative emission of nitrous oxide (N₂O) during radish and maize cropping seasons and a year with different rates of bottom-ash application.

Bottom Ash Application Rate (Mg/ha)	Cumulative N ₂ O Emission during Radish Growing Season (kg·ha ⁻¹)		Cumulative N ₂ O Emission during Maize Growing Season (kg·ha ⁻¹)		Cumulative N ₂ O Emission during Entire Year (kg·ha ⁻¹)		
	Year 1	Year 2	Year 1	Year 2	Year 1	Year 2	Year Mean
0	2.3 ^a	4.7 ^a	14.6 ^a	9.9 ^a	19.5	16.0	17.7 ^a
200	1.6 ^{ab}	2.6 ^b	9.9 ^b	4.5 ^b	12.9	8.0	10.4 ^b
400	1.0 ^b	1.5 ^c	6.3 ^c	4.0 ^b	9.2	6.9	8.0 ^c
Rate mean ¹					13.8 ^A	10.3 ^A	-

¹ Rate mean: mean value across bottom ash rates of 0, 200, and 400 Mg·ha⁻¹. Lowercase letters in the columns indicate significant differences at $p < 0.05$ for all treatments. Uppercase letters in the rows indicate significant differences at $p < 0.05$ between Years 1 and 2.

A significant R × Y interaction was noted for cumulative N₂O emission during the maize growing season (Table 3). Cumulative N₂O emission during the maize growing season increased significantly with an increasing BA application rate in Year 1 and with a BA application rate of up to 200 Mg·ha⁻¹ in Year 2 (Table 4).

Application rate, but not year, significantly influenced cumulative N₂O emission during the entire year (Table 3). The mean value of cumulative N₂O emission during the entire year across both Years 1 and 2 decreased significantly with increasing BA application rate (Table 4).

3.3. Changes in the Physical and Chemical Properties of Soil

Mean WFPS values during the radish growing season, maize growing season, and the entire year for Years 1 and 2 decreased with BA application (Table 5). Mean WFPS values during the radish growing season decreased significantly following application of 200 Mg·ha⁻¹ of BA in Year 1 and with increasing BA application rate in Year 2. Mean

WFPS values during the maize growing season and the entire year across both Years 1 and 2 decreased significantly following BA application of 200 Mg·ha⁻¹. Mean WFPS values during the maize growing season and entire year across application rates were higher in Year 1 than in Year 2, although there was no significant difference between mean values for both years.

Table 5. Averaged water-filled pore space (WFPS) of soil during radish and maize growing seasons and the entire year with different application rates of bottom ash in Years 1 and 2.

Application Rate (Mg·ha ⁻¹)	Averaged WFPS during the Radish Growing Season (%)		Averaged WFPS during the Maize Growing Season (%)			Averaged WFPS during the Entire Year (%)		
	Year 1	Year 2	Year 1	Year 2	Year Mean	Year 1	Year 2	Year Mean
0	54.9 ^a	71.0 ^a	73.1	66.6	69.9 ^a	71.9	68.5	70.2 ^a
200	34.7 ^b	53.0 ^b	55.0	51.6	53.3 ^b	53.6	52.3	52.9 ^b
400	29.3 ^b	43.3 ^c	47.3	44.7	46.0 ^b	46.1	44.5	45.3 ^b
Rate mean ¹	-	-	58.5 ^A	54.3 ^A	-	57.2 ^A	55.1 ^A	-

¹ Rate mean: mean value across application rates of 0, 200, and 400 Mg·ha⁻¹. Lower case letters in the columns indicate significant differences at $p < 0.05$ among all treatments. Upper case letters in the rows indicate significant differences at $p < 0.05$ between Years 1 and 2.

The mean value of soil pH at maize harvest time across both Years 1 and 2 increased significantly with a BA application rate of up to 200 Mg·ha⁻¹, but no further increase was observed with additional BA application (Table 6).

Table 6. Changes in chemical properties of soil amended with different application rates of bottom ash (BA) during maize harvest time.

BA Application Rate (Mg·ha ⁻¹)	pH	Electrical Conductivity (dS/m)	Organic Matter (g/kg)	Total Nitrogen (g/kg)	AV. P ₂ O ₅ (mg/kg)	Exchangeable Cations (cmol _c ·kg ⁻¹)			Bulk Density (g/cm ³)	
						K	Ca	Mg		
Year	0	5.5 ^b	0.6 ^a	18.8 ^a	1.16 ^a	277 ^a	0.57 ^a	5.38 ^a	1.77 ^a	1.17 ^a
mean	200	5.9 ^a	0.6 ^a	19.0 ^a	1.09 ^a	278 ^a	0.58 ^a	5.47 ^a	1.82 ^a	1.04 ^b
	400	5.8 ^a	0.7 ^a	19.9 ^a	1.16 ^a	285 ^a	0.58 ^a	5.44 ^a	1.82 ^a	1.01 ^b

Year mean: mean value across Years 1 and 2. Different lower case letters denote significance at $p < 0.05$ for comparisons within column.

A trade-off effect was observed between NO₃⁻ and NH₄⁺ concentrations in the soil during radish and maize growing seasons in both Years 1 and 2 (Figure 3). Soil NO₃⁻ concentration increased with decreasing soil NH₄⁺ concentration. NO₃⁻ and NH₄⁺ concentrations varied during radish and maize growing seasons in Years 1 and 2. However, NO₃⁻ concentration in soil amended with 400 Mg·ha⁻¹ of BA was always the highest, followed by that in soil amended with 200 and 0 Mg·ha⁻¹ of BA during the radish and maize growing seasons in Years 1 and 2. Conversely, NH₄⁺ concentration in soils amended with 0 Mg·ha⁻¹ of BA was always the highest, followed by that in soil amended with 200 and 400 Mg·ha⁻¹ of BA during the radish and maize growing seasons in both Years 1 and 2.

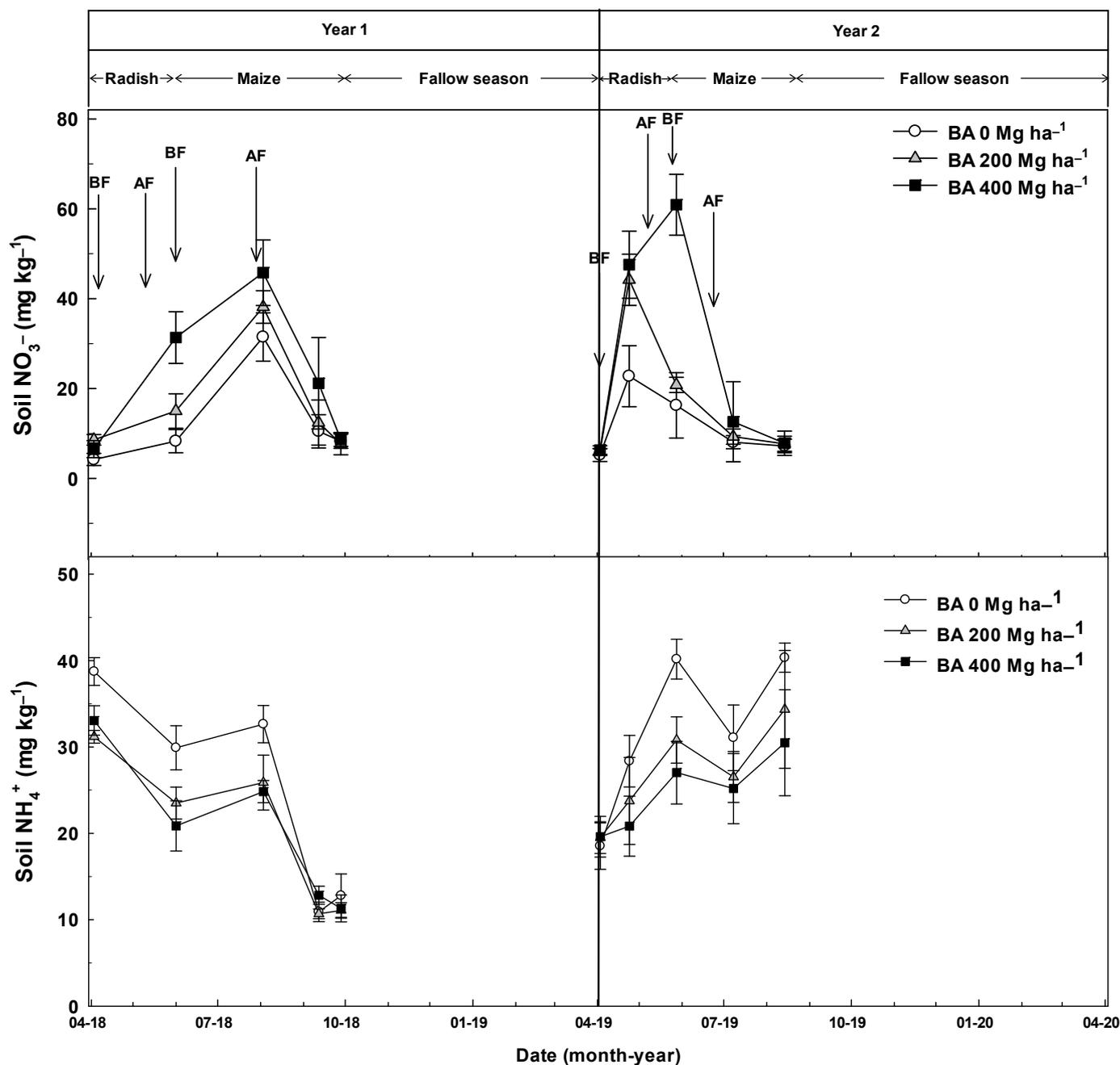


Figure 3. Seasonal changes in nitrate (NO_3^-) and ammonium (NH_4^+) concentrations in the top 15 cm soil depth following application of different rates of bottom ash during the radish and maize growing seasons in Years 1 and 2.

3.4. Crop Yield and Yield-Scaled N_2O Emission

BA application rate did not influence radish and maize yields in this study (Tables 3 and 7).

BA application rate significantly influenced yield-scaled N_2O emissions (Table 3). Radish yield-scaled N_2O emission decreased significantly with increasing application rates of BA (Table 7). Maize yield-scaled N_2O emission and total yield-scaled N_2O emission decreased significantly following a BA application rate of up to $200 \text{ Mg}\cdot\text{ha}^{-1}$, but no further decrease was noted with additional BA application.

Table 7. Biomass yields of radish and maize and yield-scaled nitrous oxide (N₂O) emission with different rates of bottom-ash application.

	Bottom Ash Rate (Mg·ha ⁻¹)	Year Mean
Radish yield	0	1.51 ^a
	200	1.43 ^a
	400	1.49 ^a
Maize yield	0	5.26 ^a
	200	5.35 ^a
	400	5.14 ^a
Radish yield-scaled N ₂ O emission (kg·N ₂ O·Mg ⁻¹ radish)	0	3.16 ^a
	200	1.90 ^b
	400	1.01 ^c
Maize yield-scaled N ₂ O emission (kg·N ₂ O·Mg ⁻¹ maize)	0	2.61 ^a
	200	1.41 ^b
	400	1.11 ^b
Total yield-scaled N ₂ O emissions (kg·N ₂ O·Mg ⁻¹ radish + maize)	0	2.49 ^a
	200	1.39 ^b
	400	0.99 ^b

Year mean: mean value across Years 1 and 2. Different lower case letters denote significance at $p < 0.05$ for comparisons among bottom ash rates.

4. Discussion

Precipitation affects N₂O emissions from arable soil. Some studies have reported that N₂O flux peaked after a high-rainfall event, which led to soil adopting an anoxic state for N₂O production via denitrification [33–36]. However, in this study, the peak of the N₂O flux did not appear after high-rainfall events, despite several occurrences of such events over the 2 years. All N₂O flux peaks appeared only during the radish and maize growing season. The peak of the N₂O flux did not appear soon after N-fertilizer application, but rather occurred 3–23 days later after urea application. Further, daily N₂O flux is associated with soil WFPS values. Nitrification is the predominant process for N₂O emission from soils with <60% WFPS, whereas denitrification is the predominant process of N₂O emission from soil with >60% WFPS [10–12]. When soil WFPS value is approximately 60%, N₂O production increases considerably owing to simultaneous nitrification and denitrification [37]. When soil WFPS value increases by >70%, soil environmental conditions favor denitrification and N₂ is emitted instead of N₂O [13]. In the present study, all N₂O flux peaks after N-fertilizer application during the growing season in both years appeared when WFPS value was approximately 65%, as shown in Figure 2a,c. For example, the peak of the daily N₂O flux did not appear following application of 0 Mg·ha⁻¹ of BA on 29 May 2018, when a basal N fertilizer was applied during the maize growing season in Year 1 because daily WFPS value was 86% on that day. Under these O₂-limited conditions, N₂ is primarily emitted instead of N₂O. However, the first and second peaks of daily N₂O flux following application of 0 Mg·ha⁻¹ of BA appeared on 21 June and 17 July after basal N-fertilizer application when daily WFPS values were 66% and 62%, respectively.

NO₃⁻ concentrations in soil increased with increasing BA application rate during radish and maize growing seasons (Figure 3), whereas that of NH₄⁺ decreased with increasing BA application rate. This implies that nitrification is the predominant N₂O-producing process rather than denitrification in BA-amended soils as BA application decreases soil WFPS values. A 35%–60% WFPS value constitutes favorable soil water conditions for nitrification. The mean WFPS value during the entire year across Years 1 and 2 decreased from 70.2% with 0 Mg·ha⁻¹ of BA to 52.9% and 45.3% with 200 and 400 Mg·ha⁻¹ of BA, respectively (Table 5).

As mentioned above, this study examined three hypotheses. The first hypothesis was that the application of porous BA decreases bulk density and WFPS value of soil to

render soil conditions unfavorable for microorganisms associated with N_2O -production processes, such as nitrification and denitrification. Results from this study confirmed the first hypothesis. The bulk density of soil at maize harvest time decreased significantly following BA application at $200 \text{ Mg}\cdot\text{ha}^{-1}$ (Table 6). This decrease in the bulk density of soil following BA application was owing to the physical properties of BA, including its high porosity and large surface area (Table 2). Subsequently, this reduced the WFPS of soil. The mean WFPS value during the entire year across both Years 1 and 2 decreased from 70.2% following application of $0 \text{ Mg}\cdot\text{ha}^{-1}$ of BA up to 45.3% with $400 \text{ Mg}\cdot\text{ha}^{-1}$ of BA (Table 5). Specifically, daily WFPS value with 200 and $400 \text{ Mg}\cdot\text{ha}^{-1}$ of BA was mostly below 35% during the entire year, except for high rainfall and irrigation events, as shown in Figure 2c. A WFPS value of 35–60% constitutes favorable soil water conditions for nitrification. However, nitrification and denitrification reduced in water-limited conditions involving a WFPS value of <35% [12,38]. BA application may ensure water-limited conditions for microorganisms involved in nitrification and denitrification and thus decrease N_2O emission from soil. Similar results with this study were observed by other researchers using ash materials, such as biochar and charcoal. Carvalho et al. [39] reported that WFPS value decreased significantly by approximately 10% following the application of $32 \text{ Mg}\cdot\text{ha}^{-1}$ of wood biochar in a bean-rice rotated cultivation system. They observed a positive correlation between N_2O fluxes and WFPS value, indicating that WFPS was a relevant soil variable related to N_2O emission. In addition, Yanai et al. [40] reported that suppressed N_2O emissions after adding charcoal stemmed from changes in WFPS values rather than the addition of Cl^- and SO_4^{2-} , which were the major anions in charcoal based on laboratory experiments. In the current study, a large decrease in WFPS value (from 70.2% to 45.3%) with BA application was observed compared with the results of other studies [39,40], owing to a greater BA application rate ($400 \text{ Mg}\cdot\text{ha}^{-1}$). Therefore, we observed a further decrease in cumulative N_2O emission by up to 54.8%, i.e., from $17.7 \text{ kg}\cdot\text{N}_2\text{O}\cdot\text{ha}^{-1}$ to $8.0 \text{ kg}\cdot\text{N}_2\text{O}\cdot\text{ha}^{-1}$.

The second hypothesis was that alkaline BA application increases soil pH, promotes reduction of N_2O to N_2 , and decreases N_2O emission. Soil pH increased following BA application (Table 6) owing to the chemical properties of BA such as presence of large amounts of CaCO_3 and CaO (Table 2). Soil pH is a primary factor influencing N_2O production and consumption processes in soil [41,42]. Several studies have reported that the abundance of nitrogen-cycling genes and the rates of nitrification and denitrification are strongly regulated by soil pH [43–46]. Notably, Nos activity is more sensitive to low pH than other reductases in denitrification [47]. Therefore, under low soil pH conditions, more N_2O is produced than N_2 [48]. The ratios of $N_2O/(N_2 + N_2O)$ showed a significant negative correlation with soil pH within the generally observed pH range of 5–8 in agricultural soils [45,48]. Increasing pH of acidic soil decreases N_2O emissions from nitrification by increasing ammonia oxidizing bacteria (AOB) gene copy numbers [49]. In this study, soil pH increased from 5.4 at $0 \text{ Mg}\cdot\text{ha}^{-1}$ of BA up to 5.9 following application of $200 \text{ Mg}\cdot\text{ha}^{-1}$ of BA (Table 6). Therefore, an increase in soil pH with BA application may help reduce N_2O emission from the soil by decreasing AOB abundance during nitrification and increasing Nos activity during denitrification. Kim et al. [21] reported that soil pH increased from 6.03 to 6.16 with $30 \text{ Mg}\cdot\text{ha}^{-1}$ of BA application in upland soil used for growing lettuce. In the current study, a greater increase in soil pH (from 5.4 to 5.9) with BA application was observed compared with their results as a higher BA rate ($200 \text{ Mg}\cdot\text{ha}^{-1}$) was applied. Some studies have reported that liming affects acidic soil by suppressing N_2O emissions. Shaaban et al. [50] observed that soil pH of two acidic soils (5.25 and 5.52) significantly increased by 7.62 and 7.77 after adding $15 \text{ g}\cdot\text{kg}^{-1}$ of dolomite [$\text{CaMg}(\text{CO}_3)_2$] and cumulative N_2O emissions decreased by up to 73.77% and 64.07% compared with the control, respectively. Barton et al. [49] also reported that increasing soil pH from 4.21 to 6.34 by liming decreased cumulative N_2O emission by 16.3%, i.e., from $1.66 \text{ mg } N_2O\cdot\text{N } \text{kg}^{-1}$ to $1.39 \text{ mg } N_2O\cdot\text{N } \text{kg}^{-1}$.

The third hypothesis was that BA comprising various elements provides available nutrients for plant growth and increases crop yield. However, this hypothesis was not confirmed based on our results. Radish and maize yields did not increase with increasing BA application rate. Rather, BA application decreased bulk density and WFPS value of soil in the present study (Tables 5 and 6), implying improved physical properties, such as porosity and aeration. However, several studies have reported that crop yield was not influenced by changes in physical soil properties for a short period of <5 years [51–54]. This study was conducted for a short period of 2 years. Improved crop yield through changes in physical properties of soil via BA application may have been observed for a study period of >5 years. In addition, macronutrient concentration, such as total nitrogen, available phosphate, and exchangeable K and Ca in soil for plant growth, did not increase with increasing BA application rates. However, BA used in this study comprised nitrogen, phosphorus, CaCO_3 , and CaO (Tables 2 and 6). We assumed that the low solubility of BA in the soil did not enrich the soil with sufficient plant nutrients to increase radish and maize yields.

In this study, daily N_2O fluxes were measured using a static chamber without growing any plants, including radish, maize, and weeds, for the entire experiment period. This may incorrectly reflect the cumulative N_2O emission from the real crop cultivation system because the effect of rhizosphere soil on N_2O emission was excluded. Rhizosphere soil encompasses the narrow zone of contact between the roots and soil particles and plays an important role for both plant growth and N_2O emissions [55,56]. Rapid nitrogen transformations and translocations occur in the rhizosphere soil via root uptake and microbial activities mainly from roots and microorganisms interacting and competing with each other for nutrients [57]. Plant roots directly capture NH_4^+ , NO_3^- , and amino acids according to their growth demands while excreting sugars, organic acids, and amino acids, which stimulate microbial growth and influence carbon and the nitrogen biogeochemical cycles. Despite the importance of the rhizosphere and its management for developing effective mitigation strategies, studies quantifying N_2O emission in the rhizosphere are surprisingly scarce owing to the lack of appropriate tools for performing measurements in such microenvironments. Measuring N_2O emission in microenvironments remains challenging owing to soil heterogeneity and the gaseous nature of N_2O [58,59].

Agricultural practices can be associated with N_2O emission by estimating cumulative N_2O emission based on crop yield, which are referred to as yield-scaled N_2O emission. From global environment and food security perspectives, sustainable agriculture in the future should explore systems with low N_2O emissions and high crop yield to ensure food security. Yield-scaled N_2O emission provides essential information for estimating the environmental impacts of intensive agricultural production systems [60]. The mean value of total yield-scaled N_2O emission across both Years 1 and 2 decreased significantly following BA application at $200 \text{ Mg}\cdot\text{ha}^{-1}$ and no further decrease was observed after additional BA application (Table 7). A lower value of yield-scaled N_2O emission indicates less N_2O emission for the same unit of crop production. Therefore, applications of up to $200 \text{ Mg}\cdot\text{ha}^{-1}$ of BA may be environmentally and economically beneficial in soil management to reduce N_2O emissions while maintaining crop production.

5. Conclusions

BA application significantly decreased cumulative N_2O emission, but did not increase crop yield. BA application of up to $200 \text{ Mg}\cdot\text{ha}^{-1}$ significantly decreased yield-scaled N_2O . Mitigating N_2O emission with BA application was mainly attributed to reduced soil WFPS value and increasing soil pH. For economic viability and environmental conservation, we recommend a BA application rate of $200 \text{ Mg}\cdot\text{ha}^{-1}$ to reduce N_2O emissions per unit of crop production.

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