

Soil N intensity as a measure to estimate annual N₂O and NO fluxes from natural and managed ecosystems[☆]

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As natural and managed terrestrial ecosystems are major sources of the potent greenhouse gas nitrous oxide (N₂O) and of the atmospheric pollutant nitric oxide (NO), predicting the source strengths of these ecosystems is central to understanding and sustainably managing the N-oxides fluxes. Here we reviewed 82 high temporal resolution datasets on N₂O and 57 on NO fluxes collected from multi-site and multi-year field measurements, including grasslands, forests, and agricultural crops, to assess whether soil N intensity, that is, the time-weighted sum of soil NH₄⁺ and/or NO₃⁻ concentrations, can be used to estimate annual N-oxides emissions. We show that soil N intensity alone can accurately predict annual N₂O and NO emissions, and that the N₂O emission strength is exponentially related to the soil inorganic N load. Thus, measuring soil inorganic N loads should improve current estimates of N-oxide emissions from global terrestrial ecosystems, and open possibilities for monitoring N₂O mitigation efforts.

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Introduction

Nitrogen (N)-oxide gases (i.e. nitrous oxide [N₂O] and nitric oxide [NO]) play an important role in the biogeochemical N cycling, but also have significant environmental impacts [1,2]. Nitrous oxide is known as a potent greenhouse gas (GHG) as well as the most abundant stratospheric ozone (O₃)-depleting substance [3], and NO affects tropospheric O₃ concentrations, an important GHG and atmospheric oxidant, and aerosols through atmospheric chemistry [4]. All of these effects contribute to global radiative forcing. Soils are recognized as major sources of both atmospheric N₂O and NO. According to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [5], soils under agricultural crops and natural vegetation contribute 5.0–13.8 Tg N₂O-N and 8.7–11.7 Tg NO-N to the atmosphere annually. Uncertainties in estimates of the source strengths of N₂O and NO are still very large, but might be narrowed if easy to measure proxies can be used to predict soil N-oxide fluxes [e.g. Refs. 6–9,10^{*}].

So far, while studies have identified empirical relationships to quantify soil N-oxide fluxes for individual sites, these relationships often fail to work for other sites [11]. Relationships that were found to be sufficiently robust to approximate emissions across sites differing widely in environmental conditions (management, vegetation, soil properties, and climate) remain rare and limited in their regional and temporal scope. For example, Rochette *et al.* [10^{*}] showed that growing season N₂O fluxes from Canadian agricultural soils can be predicted by growing season precipitation, soil texture (coarse, medium and fine), type of N (synthetic and organic), and crop type (perennial and annual), although the accuracy of this approach at larger scales remains untested.

In soils, N-oxides emissions are closely linked to the microbial processes of nitrification (the oxidation of ammonium [NH₄⁺] to nitrite [NO₂⁻] and nitrate [NO₃⁻]) and denitrification (the reduction of NO₃⁻ and NO₂⁻ to NO, N₂O, and dinitrogen [N₂]) [12,13]. Both processes use soil mineral N (NH₄⁺ and NO₃⁻) as substrates that originate mainly from the mineralization of organic matter or, in managed systems, from fertilization. Consequently, there is a strong theoretical basis for the relationship between soil

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N availability, N cycling rates and N-oxide fluxes. This context can be well described by the conceptual ‘hole-in-the-pipe’ model [14,15], within which rates of N cycling and N availability control the ‘size of the pipe’ (i.e. the amount of N that is nitrified and denitrified), while the ‘sizes of the holes’ (i.e. the N-oxide losses from each process) are controlled by factors such as soil moisture, pH and carbon content as well as the types and quantities of microbes (nitrifiers and denitrifiers). However, N cycling rates and N availability (e.g. soil net N mineralization and inorganic N concentrations) are temporally and spatially highly variable so that in most studies only weak or no correlations between measured N-oxide fluxes and soil NH_4^+ and/or NO_3^- concentrations in cropland [e.g. $R^2 = 0.08$; 16], grassland [e.g. $R^2 = 0.04$; 17*] or forest [e.g. $R^2 = 0.17$; 18] have been found.

However, this lack of correlation may be different if observations are averaged across longer time intervals, that is, at seasonal or annual time scales. The use of soil N intensity is such an approach. N intensity (i.e. the time-weighted sum of soil NH_4^+ and/or NO_3^- concentrations) is an integrated measure indicating the longer term availability of inorganic N to the soil microbial community, which is supposed to affect the presence and activity of nitrifiers and denitrifiers in soils [16]. On longer time scales, such as months to years, several studies found strong correlations between cumulative N_2O emissions and soil NO_3^- -N intensity [19–24]. However, the capacity of such an approach to scale emissions across sites was never tested. Thus, we hypothesized that soil N intensity is a proxy that can be used to accurately scale annual soil N_2O and NO fluxes across natural and managed ecosystems, and that compared to soil N intensity other soil environmental factors mostly play a minor role for predicting annual fluxes. We tested this hypothesis by screening the existing literature for datasets on annual N-oxide fluxes and measured soil inorganic N concentrations for various landuses, soil types, and climatic conditions.

Data

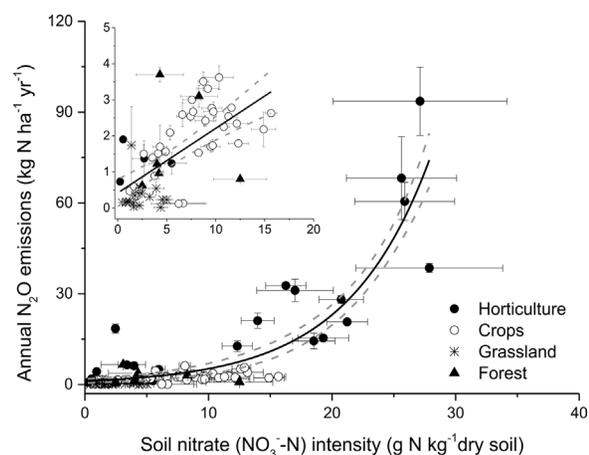
To test our hypothesis, we compiled a database of observed annual N_2O and NO fluxes and topsoil (usually 0–10 cm) inorganic N concentrations. In total, data from 13 sites were used, mainly in China, but also including sites in Africa, Europe, and Australia, comprising 82 treatment-years (see Table S1 in the Supplementary material). In all studies N_2O and NO fluxes were measured using either manual or automated chamber-based techniques with sampling frequencies varying from subdaily to weekly. In the selected studies, topsoil inorganic N concentrations were determined every 1–2 weeks. In all studies, soil inorganic N was measured by extracting soils with a salt solution (CaCl_2 , KCl, or K_2SO_4), and measuring the concentrations of NH_4^+ and NO_3^- in the extracts colorimetrically. Soil N (NH_4 -N and NO_3 -N or the sum of

both) intensity was calculated as the integrated sum of topsoil inorganic concentrations over a one year period [16]. In addition to soil N intensity, N application rate, climate (e.g. precipitation), mean soil temperature, soil organic C, clay content, and pH were used in the regression analysis. Non-linear, linear, and stepwise multiple regression analysis were used to test if a relationship between soil N intensity and/or other site characteristics and annual N_2O and/or NO fluxes exist. Statistical analyses were done using the SPSS software (SPSS China, Beijing, China).

Relationships between N intensity and annual N-oxides emissions

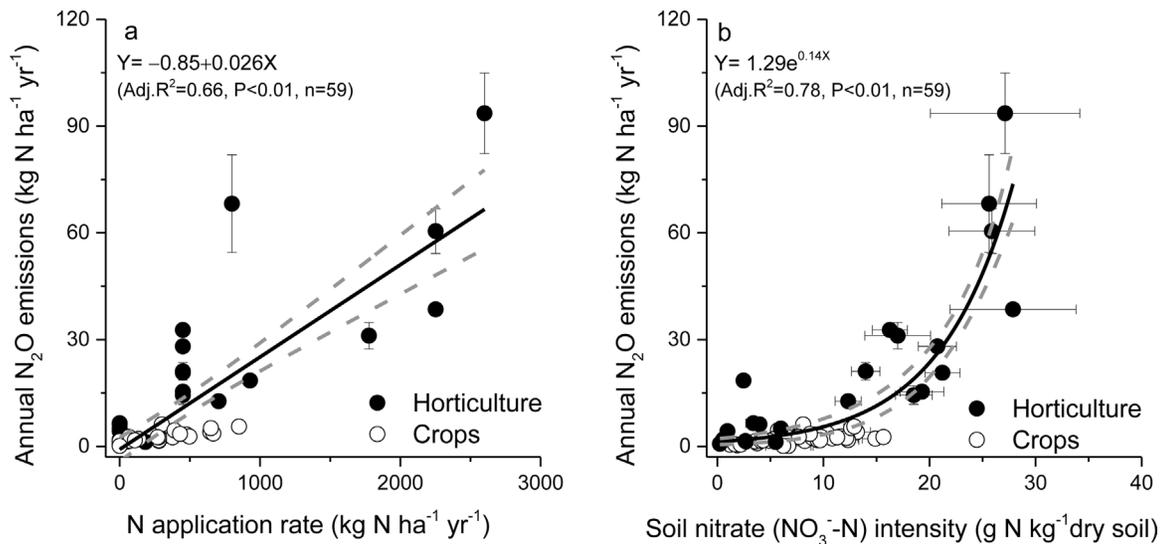
Across all sites, landuses, climate and soil conditions, stepwise multiple linear regression analysis showed that soil NO_3^- -N intensity was the most important factor controlling N_2O emissions, explaining 57% of the variation on its own and 69% of the variation when combined with four other soil/climatic variables (see Table S2 in the Supplementary material). However, an exponential model using only soil NO_3^- -N intensity explained 79% of the variation in annual N_2O emissions (Figure 1); 10% more variation than the 5-factor multiple linear regression. Correlations with individual soil properties (e.g. soil pH or clay content) alone were not significant ($P > 0.05$), but the addition of soil pH and clay content improved the exponential model with soil NO_3^- -N intensity,

Figure 1



Relationship between soil nitrate (NO_3^- -N) intensity and annual nitrous oxide (N_2O) emissions across various landuses (horticulture (i.e. tea, vegetable and orchard), different arable crops, grasslands and forests, see Table S1) ($F_{\text{N}_2\text{O}} = 1.18e^{0.15\text{NO}_3^- \text{N intensity}}$, adj. $R^2 = 0.79$, $P < 0.01$, $n = 82$). The inset highlights the relationship for sites with annual N_2O fluxes $< 4 \text{ kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ ($F_{\text{N}_2\text{O}} = 0.42 + 0.18\text{NO}_3^- \text{N intensity}$, adj. $R^2 = 0.41$, $P < 0.01$, $n = 59$). Dotted lines represent 95% confidence bands and bars represent ± 1 standard error.

Figure 2



Relationship between (a) N application rate or (b) soil nitrate (NO₃⁻-N) intensity and annual nitrous oxide (N₂O) emissions for sites receiving N-fertilizers, that is, horticulture (i.e. tea, vegetable and orchard) and arable cropping sites (see Table S1). Dotted lines represent 95% confidence bands and bars represent ± 1 standard error.

explaining 87% of the variability in annual N₂O emissions (see Figure S1 in the Supplementary material).

If the sites and observation years with annual N₂O emissions > 4 kg N₂O-N ha⁻¹ yr⁻¹ are excluded (i.e. excluding observations from intensively fertilized [>200 kg N ha⁻¹ yr⁻¹] horticultural sites such as tea, vegetable and orchard fields), the relationship between annual N₂O emissions and soil NO₃⁻-N intensity changes from non-linear to a linear relationship (Adj. R² = 0.41, P < 0.01, n = 59; Figure 1). For the agricultural fields (i.e. horticulture and different arable crops), there was also a positive correlation between N application rate and annual N₂O emissions (i.e. the IPCC approach); however, this resulted in a lower R² as compared to the relationship of annual soil N₂O emissions with soil NO₃⁻-N intensity (i.e. Adj. R² = 0.66 versus 0.78, Figure 2).

Similar to N₂O, stepwise regression analysis showed that annual NO fluxes correlated most closely with soil NH₄⁺-N intensity (Adj. R² = 0.94, P < 0.01, n = 57; see Figure 3 and Table S2 in the Supplementary material). Weaker, but significant, negative correlations were found between soil pH and annual NO emissions (Adj. R² = 0.37, P < 0.01, n = 57) and soil clay content and annual NO emissions (Adj. R² = 0.24, P < 0.01, n = 57). However, including all parameters in a multiple regression analysis did not result in an improved model as compared to the relationship with soil NH₄⁺-N intensity alone. Observations with high soil NO fluxes (> 4 kg NO-N ha⁻¹ yr⁻¹) were mainly from tea and forest sites that are typically

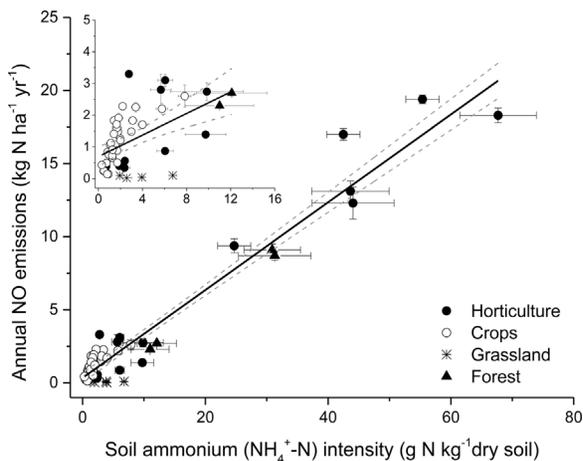
acidic [pH < 5.5; 25,26,27], which drives NO formation via chemo-denitrification rather than by nitrification [28]. Unlike with the N₂O emissions, excluding these high emitting sites did not significantly change the relationship between NH₄⁺-N intensity and annual NO emissions, but only reduced the R² value (Adj. R² = 0.28, P < 0.01, n = 49; Figure 3).

Information on soil N (NH₄⁺+NO₃⁻) intensity can also be efficiently used to predict the sum of annual N₂O + NO emissions (Adj. R² = 0.62, P < 0.01, n = 57; see Table S2 in the supplementary material and Figure 4). Similar to the NO, there were negative relationships between annual N₂O + NO emissions and both soil pH (Adj. R² = 0.18, P < 0.01, n = 57) and clay content (Adj. R² = 0.14, P < 0.01, n = 57). Total water input (precipitation plus irrigation water) was also weakly, but positively correlated to annual N₂O + NO emissions (Adj. R² = 0.13, P < 0.01, n = 57). However, the only significant parameters determined by the stepwise regression were water input and soil inorganic N intensity (Adj. R² = 0.69, P < 0.01, n = 57; see Table S2 and Figure S2 in the Supplementary material). Limiting the regression analysis to observations with annual N₂O + NO fluxes < 8 kg N ha⁻¹ yr⁻¹ did not significantly change the equation ($F_{N_2O+NO} = 0.81 + 0.27 \times \text{Inorganic N intensity}$) but only resulted in a lower R² (Adj. R² = 0.47, P < 0.01, n = 41).

Perspective and implications

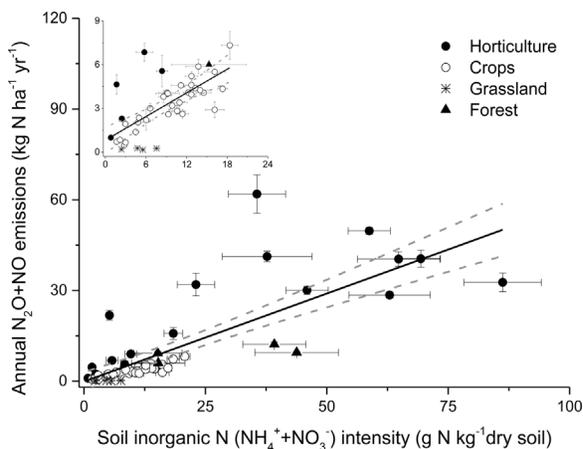
Here we show for the first time that the integrated sum of soil inorganic N concentrations and annual N₂O as well as

Figure 3



Relationship between soil ammonium ($\text{NH}_4^+\text{-N}$) intensity and annual nitric oxide (NO) emissions across sites with various landuses: horticulture (i.e. tea, vegetable and orchard), arable crops, grasslands and forests (see Table S1) (i.e. $F_{\text{NO}} = 0.35 + 0.30 \text{NH}_4^+\text{-N intensity}$, adj. $R^2 = 0.94$, $P < 0.01$, $n = 57$). The inset highlights the relationship for low NO-emitting points $< 4 \text{ kg NO-N ha}^{-1} \text{ yr}^{-1}$ (i.e. $F_{\text{NO}} = 0.69 + 0.17 \text{NH}_4^+\text{-N intensity}$, adj. $R^2 = 0.28$, $P < 0.01$, $n = 49$). Dotted lines represent 95% confidence bands and bars represent ± 1 standard error.

Figure 4



Relationship between soil inorganic N ($\text{NO}_3^- + \text{NH}_4^+$) intensity and annual $\text{N}_2\text{O} + \text{NO}$ emissions across various landuses (horticulture (i.e. tea, vegetable and orchard), arable crops, grasslands and forests in Table S1) ($F_{\text{N}_2\text{O}+\text{NO}} = -0.10 + 0.58 \text{Inorganic N intensity}$, adj. $R^2 = 0.62$, $P < 0.01$, $n = 57$). The inset highlights the relationship for low $\text{N}_2\text{O} + \text{NO}$ -emitting points $< 8 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ($F_{\text{N}_2\text{O}+\text{NO}} = 0.81 + 0.27 \text{Inorganic N intensity}$, adj. $R^2 = 0.47$, $P < 0.01$, $n = 41$). Dotted lines represent 95% confidence bands and bars represent ± 1 standard error.

NO emissions are closely related across a wide variety of sites, landuses, soil conditions, climate and, thus, soil environmental conditions. Such a relationship had been proposed earlier [14,15], although previous studies have

mostly failed to demonstrate a close link between soil inorganic N concentrations and instantaneous in-situ N_2O fluxes. The lack of correlation between instantaneous fluxes and inorganic N concentrations is because denitrification, which dominates N_2O production in agricultural systems [29,30], and requires the combination of anaerobic conditions and the presence of an electron donor as well as a source of NO_3^- . Nitrous oxide may also be produced by nitrification, which tends to vary less over time; however, the product ratio for nitrification is very low [0.02 to 0.09%; 31] compared to denitrification [$\sim 9\%$; 2,32]. As a result, high NO_3^- concentrations alone will not produce high N_2O fluxes. But unlike many previous studies, we did not attempt to link seasonal dynamics or short-term emission peaks to soil inorganic N variations, but rather used aggregated annual values [16,19–21]. The use of aggregated inorganic N values tends to correspond better with annual emissions because the probability of anaerobic conditions occurring concurrently with sufficient NO_3^- increases with greater N intensities, likely resulting in more episodes of denitrification. This approach is similar to the IPCC approach [33] that estimates soil N_2O emissions based on fertilizer inputs. The non-linear relationship between soil NO_3^- intensity and N_2O emissions though, seems to be as robust as the IPCC approach (Figure 2) while better describing the exponential increase of soil emissions of highly fertilized soils [34–37]. Unlike the IPCC approach though, soil NO_3^- intensity can also be used to predict fluxes from non-fertilized systems.

The strong exponential correlation between soil NO_3^- intensity and cumulative annual N_2O fluxes also suggests that reducing soil NO_3^- concentrations is key to mitigating N_2O fluxes. Thus, utilization of the 4R fertilization strategy [38,39], which matches fertilizer type, application rate, location and timing to the plant needs should reduce N_2O emissions even if fertilizer N application rates remain high. Also, the utilization of plant species that can improve N use efficiency [e.g. using plant trait-based approaches such as species mixtures for grasslands and cover crops, intercropping systems, diversified crop rotations and breeding programs, 40**] or inhibit nitrification [e.g. through biological nitrification inhibition – BNI, 41] should also reduce soil NO_3^- intensities and hence N_2O fluxes as well.

The addition of soil property information (soil texture and pH in the case of N_2O) into the model further improved the accuracy of the estimates, explaining 87% of the observed variability (see Figure S1). As information on soil inorganic N concentration is easier to gather than field GHG flux measurements, and as recent advancements in sensor technologies that quickly and directly measure inorganic N concentrations in the plant rooting zone come online [42*], the approach suggested here should improve estimates of regional N_2O emissions. This could be

especially useful for estimating anthropogenic N₂O emissions from agriculture, forestry and other land-use activities that do not include fertilizer applications (e.g. landuse change, forest degradation due to human activity/N saturation/pollution, etc.). Furthermore, this does not only hold for N₂O, but also for NO emissions from soils, which are a significant source for regional tropospheric NO_x concentrations.

In summary, the empirical models developed in this study explain 79% of the variance in annual N₂O emissions from natural and managed ecosystems, and 94% of the variance in annual NO fluxes. These models are innovative in that the strong link between aggregated inorganic N concentrations and annual N-oxide emissions will allow us to use soil inorganic N data to support mitigation policy and produce national inventories. In future studies, the reliability of these models should be verified using data external to that used to build the model and improved by using additional datasets with better geographical representation and temporal distribution.

Conflict of interest statement

Nothing declared.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.cosust.2020.03.008>.

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- of outstanding interest

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