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Skiba, U., Medinets, S., Cardenas, L. M., Carnell, E. J., Hutching, N. J. and Amon, B. 2021. Assessing the contribution of soil NOx emissions to European atmospheric pollution. *Environmental Research Letters*. 16 (2), p. 025009. https://doi.org/10.1088/1748-9326/abd2f2

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Assessing the contribution of soil NO$_x$ emissions to European atmospheric pollution

To cite this article: U Skiba et al 2021 Environ. Res. Lett. 16 025009

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Assessing the contribution of soil NO\textsubscript{x} emissions to European atmospheric pollution

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Keywords: nitric oxide, emission factor, agriculture, soil, manure, forest

Abstract

Atmospheric NO\textsubscript{x} concentrations are declining steadily due to successful abatement strategies predominantly targeting combustion sources. On the European continent, total NO\textsubscript{x} emissions fell by 55% between 1990 and 2017, but only modest reductions were achieved from the agricultural sector; with 7.8% from 20 Eastern European countries and 19.1% from 22 Western European countries. Consequently, the share of agricultural NO\textsubscript{x} emissions for these 42 European countries have increased from 3.6% to 7.2%. These values are highly uncertain due to serious lack of studies from agricultural soils and manure management. The emission factor (EF\textsubscript{NO} 1.33%), currently used for calculating soil NO\textsubscript{x} emissions from European agricultural categories ‘N applied to soils’ and ‘manure management’ was evaluated here by including recently published data from temperate climate zones. The newly calculated EF\textsubscript{NO} (average 0.60%, 0.06\textsubscript{25th}/0.54\textsubscript{75th}%, \(n = 65\) studies) is not notably different from the current value, given the large uncertainties associated with the small pool of studies, and therefore continued use of EF\textsubscript{NO} (1.33%) is recommended until more data become available. An assessment of the contribution of agricultural and non-agricultural NO\textsubscript{x} sources found that of the 42 European countries, the 8 most populated countries achieved considerable reductions (1990–2017) from categories ‘non-agricultural sources’ (55%), ‘N applied to soils’ (43%) and ‘manure management’ (1.2%), compared to small reductions from the remaining 34 countries. Forests are also large sources of soil NO\textsubscript{x}. On average, emissions from Eastern European forests were 4 times larger than from ‘N applied agricultural soil’, whereas Western European NO\textsubscript{x} emissions from ‘N applied agricultural soil’ were two times larger than from forest soils. Given that non-agricultural sources of NO\textsubscript{x} continue to decline, soil related emissions from agriculture, forests and manure management become more important, and require rigorous investigation in order to improve atmospheric pollution forecasts.

1. Introduction

Nitric oxide (NO) is a main component of the nitrogen oxides (NO\textsubscript{x}: NO + NO\textsubscript{2}), a highly reactive and short-lived molecule, involved in the tropospheric chemistry of photochemical production of ozone (O\textsubscript{3}) and aerosols, impacting on human health, global warming, crop production and eutrophication of vulnerable ecosystems.

Globally, combustion sources (fossil fuel, biomass and biofuel burning and wildfires) account for 76% of total NO\textsubscript{x} emissions. Soils contribute 20% (natural soils account for 12% and agricultural soils for 8%), and aviation and lightning around 4% [1, 2].

It is estimated that 4%–8% of the total EU 27 annual NO\textsubscript{x} emissions originate from nitrogen (N) fertilised agricultural soils [3]. On hot summer days, however, NO\textsubscript{x} emissions from N fertilised soils may contribute up to 27% of total NO\textsubscript{x} emissions [4]. Soil NO\textsubscript{x} emissions arising from N fertilisation are of particular importance in rural areas, where traffic and stationary combustion sources are small,
so agricultural emissions play an important role in rural air pollution, especially O\textsubscript{3} production [5, 6]. Increased O\textsubscript{3} concentrations are known to reduce crop production and forest growth [7].

In soils, NO is produced microbially and abiotically. The main microbial pathways responsible for NO production are nitrification and denitrification [8] and other recently discovered processes [6, 9]. The same groups of microorganisms involved in nitrous oxide (N\textsubscript{2}O) production are also responsible for NO emissions and uptake. Key drivers, which influence soil NO and N\textsubscript{2}O emission rates are: (a) the nature and magnitude of the N source, i.e. fertilisers, manures, mineralisation of organic matter, atmospheric N deposition; (b) the degree of soil aeration, which is controlled by soil texture, bulk density, precipitation and microbial activity; (c) temperature, which determines the rate of enzymatic reactions [10]; and (d) soil pH, which influences the role of abiotic and biotic NO production pathways [6].

Largest soil NO emissions occur in dry, well-drained and low organic matter content soils and the opposite is the case for N\textsubscript{2}O [11], although increasing the N supply and temperature increases the production of both gases. Abiotic production may occur in soils with high organic matter content and acid soils [12]. The importance of abiotic NO production relative to microbial production is largely unknown, and poorly investigated [9, 13]. Concerted efforts in Europe and many other countries have led to large reductions in NO\textsubscript{x} emissions from combustion sources. By 2017, the EU28 countries achieved an overall 58% reduction compared to 1990 levels [14]. Emissions from the agricultural sector also declined, but at much lower rates [14]. If this trend continues, the contribution of soil NO\textsubscript{x} emissions from the agricultural sector will become a major source. For example, in California significant reductions of combustion sources have led to soil NO\textsubscript{x} emissions being the major source of atmospheric NO\textsubscript{x} pollution [15].

The aim of this paper is to (a) evaluate current approaches to calculate soil NO\textsubscript{x} emissions, and (b) investigate changes in total and soil NO\textsubscript{x} concentrations for the European continent.

2. Methods

2.1. Calculation of soil NO emission rates and emission factors (EFs) for temperate climate zones

In this paper we will use the term 'NO' for microbial processes and EFs, and the general term 'NO\textsubscript{x}' when comparing different categories of NO\textsubscript{x}, which are presented as NO\textsubscript{2}, as prescribed by the European Environment Agency (EEA) [3].

Soil NO fertiliser/manure induced emission factors (EF\textsubscript{NO}) and emission rates were calculated from a global meta-analysis for the period 1988–2016 [16].

EFs for NO-N are derived from the percentage of soil NO-N emission rates in relation to fertiliser N application rates, and here labelled as EF\textsubscript{NO}. Liu’s dataset was filtered for temperate climate zones and managed crop and grasslands. Other relevant data-sets, not included by Liu et al. [16] were added [17–23]. A total of 43 studies were selected and provided 129 data from the European continent (n = 49), North America (n = 30), Japan (n = 43) and China (n = 7). Rates and categories of N fertilisers and cumulative soil NO fluxes were available from crops (n = 102), grasslands (n = 27), and background emissions (zero N application, n = 27) (table 1). Most studies applied synthetic N fertilisers (n = 98), such as urea, ammonium nitrate and ammonium sulphate with and without other N compounds, nutrients and nitrification inhibitors. Only 31 data sets included manures (i.e. cattle, pig, poultry manures, treated and untreated slurries). Synthetic N fertiliser application rates >400 kg N ha\textsuperscript{-1} were excluded; as such high rates are not widely used by farmers. The data are from plot experiments, investigating different treatments and from observations. They range from 20 to 570 d, with an average study period of 176 d. In addition, six short-term data (20, 21 d) were included as they specifically focused on the response of soil NO emissions, before and after N fertilisation [18–23]. Many of these studies also measured soil N\textsubscript{2}O emissions (n = 103), of which 73 data were fertilised with synthetic N and 30 with manures. These N\textsubscript{2}O data were included in the overall data analysis. In the case of NO, the EF\textsubscript{NO} were calculated as a percentage of the N input, with and without background emissions.

In order to compare soil NO emissions from crop and grasslands with those from forest soils, we calculated forest soil NO emission rates based on published data from temperate climate forests (n = 30) [16, 24, 25].

2.2. Calculating the contribution of soil NO\textsubscript{x} emissions to total NO\textsubscript{2} emissions for the European continent

Time series of agricultural and non-agricultural NO\textsubscript{x} emissions were obtained for the 42 European countries and divided into East/West geographical regions. Western Europe includes the former EU15 countries plus Cyprus, Iceland, Lichtenstein, Malta, Monaco, Norway and Switzerland (n = 22). Of the 24 Central and East European countries Armenia, Azerbaijan, Georgia and Kosovo were excluded (n = 20) [26]. Officially reported NO\textsubscript{2} emissions (using the 2014 reporting guidelines) were downloaded for the years 1990–2017 from the EMEP database and using GNFR (Gridding nomenclature for reporting) reporting levels [26]. The categories used were: (a) ‘K\textsubscript{AgriLivestock}, which is manure management from dairy, sheep, pigs, and other farm animal manures; (b) ‘L\textsubscript{AgriOther}, includes synthetic fertilisers, animal manures, and organic
fertilisers applied to soils, crop residues, indirect emissions; but also non-soil related activities, such as off-farm storage, field burning of residues and pesticides. These non-soil related sources are estimated to represent ~3% of total ‘L_AgricOther’ emissions in Europe. It was not possible to separate these minor sources, as they are reported as a single emission source; (c) the remaining GNFR categories include industrial sources, transport shipping, fugitive emissions and solvents. For simplicity, in this paper these three categories are labelled as (a) ‘Manure management’, (b) ‘N application to soils’, and (c) ‘Non-agricultural sources’.

Gridded NO\textsubscript{x} emission estimates (at a spatial resolution of 0.01 × 0.01 degree), as used in the EMEP model, were downloaded for the years 1990–2017 from the EMEP website and mapped for the European continent. The proportion of emissions associated with agricultural activity were estimated by comparing emissions from manure management and synthetic and organic N application to soils (GNFR K&L) against total gridded emissions. As N application to soils is a key driver of NO\textsubscript{x} emissions, country level data of FAO domains ‘synthetic fertilisers’ and ‘manure application to soils’ were downloaded from FAOSTAT [27] for the years 2002 and 2017, and linked to European country boundary information for 2016 [28]. European population statistics were downloaded from [29], and areas of agricultural and forest land use were downloaded from FAOSTAT [27].

### 3. Results

#### 3.1. Soil NO and N\textsubscript{2}O emission rates and factors for temperate climate zones

The average rate of synthetic N fertilisers and manures applied to crop and grasslands across 129 studies in temperate climates (140 ± 6.9 kg N ha\textsuperscript{-1}) resulted in average cumulative NO-N and N\textsubscript{2}O-N emissions of 1.37 ± 0.34 kg NO-N ha\textsuperscript{-1} and 3.86 ± 0.56 kg N\textsubscript{2}O-N ha\textsuperscript{-1}, respectively; and of 0.18 ± 0.06 kg NO-N ha\textsuperscript{-1} and 0.95 ± 0.27 kg N\textsubscript{2}O-N ha\textsuperscript{-1}, when background emissions (no N applied) were subtracted [16–23]. Disaggregation to crop and grasslands resulted in 2.6 times larger soil NO emissions from crops than grasslands. The opposite was the case for soil N\textsubscript{2}O emissions, with 1.3 times larger N\textsubscript{2}O emissions from grasslands (figure 1). The type of synthetic N fertilisers and manures applied to crop and grasslands did not significantly influence soil NO emission rates. However, soil N\textsubscript{2}O emissions after manure applications were four times larger from crops, and three times larger from grasslands compared to synthetic N fertilisers (figure 1).

Soil NO EFs were calculated without ‘(NO)’ and with background (Bg) emissions ‘(NO-Bg)’.

**Table 1.** Soil NO emission factors (EF\textsubscript{NO}) without background emissions (‘NO’) and with background emissions (‘NO-Bg’) for synthetic and/or manure N applications for croplands and/or grasslands were calculated from cumulative NO emissions (kg NO-N ha\textsuperscript{-1}) and N application rates (kg N ha\textsuperscript{-1}), using temperate climate data from Liu et al [16], and see section 2.1.

|                | Synthetic and Manure N | Synthetic N | Manure N |
|----------------|------------------------|-------------|----------|
|                | NO | NO-Bg | NO | NO-Bg | NO | NO-Bg |
| **Cumulative NO-N emissions as a % of N applied** | | | | | | |
| **Croplands and Grasslands (all data)** | | | | | | |
| Average | 1.40 | 0.60 | 1.51 | 0.49 | 1.04 | 1.01 |
| Median | 0.40 | 0.28 | 0.47 | 0.35 | 0.29 | 0.07 |
| SE\textsuperscript{a} | 0.26 | 0.19 | 0.32 | 0.08 | 0.43 | 0.87 |
| Min\textsuperscript{b} | 0.00 | 0.01 | 0.00 | 0.01 | 0.02 | 0.01 |
| Max\textsuperscript{c} | 18.74 | 12.28 | 18.14 | 2.58 | 12.66 | 12.28 |
| n\textsuperscript{d} | 129 | 65 | 98 | 51 | 31 | 14 |
| **Croplands** | | | | | | |
| Average | 1.56 | 0.55 | 1.67 | 0.31 | 1.19 | 1.57 |
| Median | 0.40 | 0.15 | 0.45 | 0.27 | 0.29 | 0.05 |
| SE | 0.33 | 0.28 | 0.39 | 0.05 | 0.57 | 1.53 |
| Min | 0.00 | 0.01 | 0.00 | 0.01 | 0.02 | 0.01 |
| Max | 18.74 | 12.28 | 18.74 | 1.08 | 12.66 | 12.28 |
| n | 102 | 43 | 79 | 35 | 23 | 8 |
| **Grasslands** | | | | | | |
| Average | 0.79 | 0.71 | 0.86 | 0.87 | 0.62 | 0.26 |
| Median | 0.47 | 0.47 | 0.48 | 0.54 | 0.29 | 0.21 |
| SE | 0.16 | 0.15 | 0.19 | 0.19 | 0.28 | 0.08 |
| Min | 0.13 | 0.03 | 0.13 | 0.10 | 0.17 | 0.03 |
| Max | 2.85 | 2.58 | 2.85 | 2.58 | 2.53 | 0.53 |
| n | 27 | 22 | 19 | 16 | 8 | 6 |

\textsuperscript{a}SE = standard error of the mean. 
\textsuperscript{b}Min = minimum. 
\textsuperscript{c}Max = maximum. 
\textsuperscript{d}n = number of observations.
Including background emissions reduced the number of data points from 129 to 65 observations for synthetic N and manure N applications, and separately for applications of ‘synthetic N’ and ‘manure N’, only (table 1). Subtracting background NO emissions from synthetic and manure N data reduced the EF$_{NO}$ from 1.4% to 0.6% (table 1). Similar reductions were calculated for croplands. This is not surprising as 80% of the data are from fertilised croplands but only 20% from fertilised grasslands. In addition, 76% of the data received synthetic N fertilisers and only 24% received manures. This means the EF analysis is heavily biased towards croplands fertilised with synthetic N compounds. For croplands, EF’s without subtracting background emissions (column ‘NO’; table 1), (average EF$_{NO}$ 1.56%, range 0.00–18.74, $n = 102$) were twice as large as those from grasslands (average EF$_{NO}$ 0.79%, range 0.13–2.85, $n = 27$). However, such differences were not observed when subtracting the background EF$_{NO}$ from croplands (EF$_{NO}$ 0.55%), and grasslands (EF$_{NO}$ 0.71%) (column ‘NO–Bg’; table 1). Given the small number of data points and large min-max ranges and standard errors (SEs), we do not recommend to use separate EFs for the four individual categories: synthetic N crop/grass and manure N crop/grass (table 1). Instead, with the existing data a single EF for croplands and grasslands combined is the most statistically ‘robust’ recommendation for N fertiliser and manure induced soil NO emissions in temperate climates of EF$_{NO}$ 1.4%, (0.11%/1.0%, $n = 129$), and soil NO–Bg EF$_{NO}$ 0.6% (0.06%/0.54%, $n = 65$; data are average EF$_{NO}$ and 25th/75th percentiles).

### 3.2. Relative changes of total and soil NO$_x$ emissions across Europe

Between 1990 and 2017 non-agricultural NO$_x$ sources decreased by 51% in Eastern and 61% in Western Europe. Over this period, agricultural emissions (synthetic and manure N applications) only achieved modest reductions of 7.8% and 19.1% for Eastern and Western Europe, respectively (table 2). The faster decline of non-agricultural sources compared to agricultural sources has led to a consistent increase in the contribution of agricultural NO$_x$ emissions from 3.2% in 1990 to 5.9% in 2017 in Eastern Europe, and from 3.8% in 1990 to 7.4% in 2017 in Western Europe (figure 2).

Total NO$_x$ emission reductions between 1990 and 2017 vary significantly for individual countries, with largest reductions from the Ukraine, UK, and Czech Republic (range 73%–78%), smallest reductions from Cyprus, Norway, Serbia and Poland (range 13%–26%), and increased emissions from Montenegro (83%) and Albania (5%). The eight most populated countries, Poland, Russian Federation,
Table 2. Comparison of NO\textsubscript{x} concentrations for agricultural and non-agricultural sources in Eastern and Western European countries.

\begin{tabular}{llll}
\hline
Source of NO\textsubscript{x} (Gg NO\textsubscript{x} y\textsuperscript{-1}) & \multicolumn{3}{c}{Year} \\
& Manure Management\textsuperscript{a} & N application to soils\textsuperscript{b} & Non-agricultural sources\textsuperscript{c} \\
\hline
Eastern European countries & 1990 & 27.8 & 296.0 & 9692.6 \\
& 2005 & 13.2 & 168.7 & 6188.6 \\
& 2017 & 11.7 & 286.8 & 4786.3 \\
Western European countries & 1990 & 20.8 & 533.5 & 14406.3 \\
& 2005 & 18.7 & 427.7 & 9872.6 \\
& 2017 & 18.3 & 430.4 & 5635.1 \\
\hline
\end{tabular}

\textsuperscript{a}EMEP categories 'K_AgrLivestock.'
\textsuperscript{b}'L_AgrOther'.
\textsuperscript{c}Non-agricultural sources [26].

Figure 2. Changes in the contribution of agricultural and non-agricultural sources of NO\textsubscript{x} in Eastern and Western European countries for the period 1990–2017 [14].

Ukraine, France, Germany, Italy, Spain and the UK, accounted for 76% of the total population and for 73% of total NO\textsubscript{x} emissions of the European continent in 2017. For these eight countries, large reductions of 74% for the non-agricultural sources, with little changes for the category ‘N application to soil’ (+1.7%) and a 167% rise in livestock-related NO\textsubscript{x} emissions has shifted the balance of the dominance of non-agricultural sources in 1990 (84%) to almost equal shares of agricultural (44%) and non-agricultural (56%) emissions in 2017 (table 3). For the remaining 36 European countries, the
Table 3. Comparison of agricultural and non-agricultural NO\textsubscript{x} emissions from the 8\* most populated countries with the remaining 36 countries on the European continent.

| Year | Source of NO\textsubscript{x} (Gg NO\textsubscript{x} y\textsuperscript{-1}) | Fraction of each category (% of total) |
|------|-------------------------------------------------|--------------------------------------|
|      | Manure management\textsuperscript{b} | N application to soils\textsuperscript{c} | Non-agricultural sources\textsuperscript{d} |
| All-minus eight countries | 1990 | Gg 16.3 | 341.9 | 6229.8 |
| | 1990 | % 0.25 | 5.19 | 94.6 |
| | 2017 | Gg 10.9 | 265.8 | 2781.0 |
| | 2017 | % 0.36 | 8.69 | 91.0 |
| Eight high population countries | 1990 | Gg 36.8 | 3533.3 | 17 869.1 |
| | 1990 | % 0.17 | 16.5 | 83.3 |
| | 2017 | Gg 98.4 | 3592.2 | 4597.9 |
| | 2017 | % 1.19 | 43.3 | 55.5 |

\textsuperscript{a}Poland, Ukraine, The Russian Federation, Spain, Italy, France, UK, Germany account for 76% of the population on the European continent in 2017 [29].

\textsuperscript{b}EMEP categories 'K_AgriLivestock'.

\textsuperscript{c}'L_AgriOther'.

\textsuperscript{d}Non-agricultural sources [26].

Figure 3. Changes of synthetic N fertiliser and manure application (left map, FAO.FAOSTAT. License: CC BY-NC-SA 3.0 IGO); and agricultural NO\textsubscript{x} emissions as a proportion of total NO\textsubscript{x} emissions (right map) for 22 Western and 20 Eastern European countries over the period 2002–2017 [14, 26–28].

The contribution of the categories ‘manure management’, ‘N application to soil’ and ‘non-agricultural sources’ has hardly changed over the period 1990 and 2017. There is a slight shift from the non-agricultural to the agricultural sources, but insignificant compared to changes observed for the ‘eight high population countries’.

An additional source to include in this analysis is soil NO emissions from forests. Median fluxes from 30 forests were 0.43 kg NO-N ha\textsuperscript{-1} y\textsuperscript{-1} (range 0–9.3), and converted to NO\textsubscript{2}, 1.41 kg NO\textsubscript{2} ha\textsuperscript{-1} y\textsuperscript{-1} [16, 24, 25]. Multiplying this emission rate by forest land areas, suggests that the large areas of Eastern European forests (877 million ha in 2017) emitted 1239 Gg NO\textsubscript{2} in 2017, whereas the much smaller forested areas in West European countries (137 million ha) emitted 194 Gg NO\textsubscript{2} y\textsuperscript{-1}.

4. Discussion

4.1. Soil NO\textsubscript{x} emission rates and factors for temperate climate zones

Calculations of soil NO emission rates and fertiliser/manure induced EF’s are based on field
measurements, which investigate the response of NO-N flux to N fertiliser application rate. Ideally, the duration of these measurements should be for at least one year, to encompass seasonal differences; and even for several years to account for climate extremes. They should be of high frequency, to take into account environmental and agricultural management changes [17, 30] and of high replication rate, due to large spatial and temporal variabilities [17, 31]. They should also include control plots without N application (i.e. background emissions), preferably cover not only cropping seasons but also pre- and post-harvest periods, and account for legacy effects of previous managements (i.e. legumes, crop residues) [17]. Only a few studies have achieved this rigor for soil NO flux measurements [17, 32], compared to large numbers of such studies for N2O flux measurements [33]. There are two reasons for this discrepancy: (a) the agricultural sector is only a small source of NO (figure 1), but the main source of N2O [34] and (b) soil NO measurements are rather cumbersome, as the short lifetime of NO (30 s) requires immediate analysis with currently rather power-hungry instruments situated in the field [17, 35–37]. In contrast, N2O is a very stable gas and can be analysed off-line.

Several global meta-analyses have calculated soil EFNO, the most recent ones by Stehfest et al [38] and Liu et al [16]. The Stehfest analysis is based on 189 measurements published between 1988 and 2004. Liu et al [16] extended this data series to 2015, with 520 field measurements. Stehfest et al [38] calculated an EF for NO-N of 0.55% of N input with a 95% confidence interval of −80% and +406% for crop and grasslands (excluding legumes). Liu et al [16] provided an EFNO of 0.87% (range 0.47–1.27) for grasslands \((n = 32)\) and 0.84% (range 0.31–1.37) for croplands \((n = 113)\), and an overall global EFNO of 1.14%. The EFNO calculated in this study, using data from temperate climates (EFNO = 0.60%, range 0.01–12.28 from crop and grasslands; table 1) is not dissimilar to Stehfest’s and Liu’s calculations for global data [16, 38], considering the large uncertainty ranges. However, the EFNO (1.33%), recommended by the EEA [3], is considerably larger than the EFNO calculated in this study. The EEA EFNO (1.33%) calculation is also based on Stehfest’s compilation of global data [38], but only includes data from North America, Europe and the former USSR. For EEA inventory reporting NO emission rates and EFNO are required to be converted to NO2 in order to compare the soil NO-N emissions with combustion sources (NOx). The EFNO 1.33% translates to EFNO2 4.14% (0.04 kg NO2/kg N applied, with 95% confidence intervals of 0.005–0.104 soil NO2 emissions) [3].

The large uncertainty ranges for all four EFNO calculations (Liu, Stehfest, EEA and this study) imply that respective EFNO values (~0.87% [3], 0.55% [38], 1.33% [3], 0.60% this study) are probably not very different. We therefore decided to use the EEA EFNO to calculate the NO emission rates presented in figures 2 and 3 and tables 2 and 3.

Out of the 43 publications included in our EFNO analysis [16, 18–23], only 19 studies are post 2004. This is rather concerning, and points to the need of many more field based soil NO measurements across the European climate zones and their main agricultural management practices (crop categories, fertiliser practices, tillage, irrigation, and manure management).

Manure management EF’s (reported as NO2) were recently updated for the national reporting framework (NRF) source categories, and range from 0.0001 (laying hens: slurry) to 0.752 (dairy cattle: solids) [39]. As for soil NO emissions, these values also are highly uncertain due to lack of data. The contribution of manure management to total European NOx emissions is 0.23% in Eastern Europe and 0.30% in Western Europe in 2017 compared to 5.6% and 7.1% from synthetic and manure N applied to soils for Eastern and Western Europe, respectively (figure 2). However, per area basis manure stores are small compared to fields, but they provide concentrated hot-spots of NOx, not to be ignored.

Tier 2 NO EFs, based on, i.e. different agricultural and manure management practices, fertiliser regimes, crop and soil categories, are not available, due to the lack of sufficient data [3]. However, Tier 3 process-based modelling, using the denitrification decomposition model (DNDC), was developed to calculate soil NOx emissions from not only agricultural soils but also from forest soils (e.g. [40–42]). For the EU15 countries in the year 2007, emissions from N fertilised soils were 332 Gg NOx y−1 [5], and are in agreement with the EMEP Tier 1 approach (306 Gg NOx y−1). Considering the large uncertainties of such estimates provides some level of confidence in the EMEP EEA Tier 1 approach. One must not forget that forests are important sources of soil NOx emissions, especially in areas with large atmospheric N deposition rates. Forests are efficient traps for pollutants, with N deposition rates much larger than to short vegetation [43]. In addition the typical low forest soil bulk densities favour NO production [16, 25, 40, 44]. At the global scale forest soil NOx emission rates (2.7 kg NOx ha−1 y−1) are of the same order of magnitude as fertilised crops and grasslands (2.8 and 2.4 kg NOx ha−1 y−1) [16]. Similarly, for the EU15 agricultural emissions were only 1.3 times larger than forest emissions [5]. Pledges and policies of increased tree planting in Europe as a panacea to combat climate change may increase soil NOx emission from forests.

4.2. Relative changes in total and soil NOx emissions across Europe

On average, non-agricultural NOx emissions have decreased between 1990 and 2017 from the 42 countries of the European continent by 56%.
(figure 2, table 1), and are highly likely to decline further through developments of green transport policies and further industrial reductions [45, 46]. Agricultural sources of NO\textsubscript{x} also declined, albeit at modest rates. For Western Europe, the reduction in ‘N applied to soil’ coincided with a 4% reduction in N fertiliser application. In contrast, N fertiliser use increased by 57% in Eastern Europe (figure 3, table 2), with little change in soil NO\textsubscript{x} emissions between 1990 and 2017.

This study has provided an overview of changes in agricultural NO\textsubscript{x} emissions in response to N fertilisation and manure management within the context of total NO\textsubscript{x} emissions and focus on the collective Eastern versus Western European countries. There is large variability in the percentage contribution of agricultural soil NO\textsubscript{x} emissions to total emissions for individual countries (figure 3). For example, country level disaggregation of the change in N fertiliser application does not directly match the changes in agricultural NO\textsubscript{x} emissions as a proportion of total NO\textsubscript{x} emissions, because of a range of drivers; mainly land management, population densities and industrialisation (figure 3).

The downward trend of overall NO\textsubscript{x} emissions is encouraging, but the increased contribution from agricultural and forest soils and manure management is of concern, for several reasons. (a) Uncertainties of the soil and manure management related emissions are too large to provide accurate predictions of high O\textsubscript{3} events. Especially biogenic NO\textsubscript{x} emissions from N applications to soils, manure management and forests can be important sources of European atmospheric O\textsubscript{3} concentrations, particularly in spring (N fertilisation period) and summer periods (dry soils, which promote NO over N\textsubscript{2}O production) [47, 48]. (b) Climate change predictions of reduced rainfall and increased drought events across many parts of Europe will favour soil NO\textsubscript{x} over soil N\textsubscript{2}O emissions from N fertilised soils, because dry and well aerated soils provide the O\textsubscript{2} required for microbial NO production, whereas low O\textsubscript{2} concentrations are needed for N\textsubscript{2}O production [6, 8]. (c) Increased tree planting may increase soil NO\textsubscript{x} emissions unless atmospheric N deposition rates arising from agricultural reactive N emissions (mainly ammonia) and fossil fuel combustion sources are declining. A potential NO\textsubscript{x} mitigation option would be to lime forest soils, but this could increase N\textsubscript{2}O emissions and NO\textsubscript{3} leaching, which needs to be studied prior lime application to avoid pollution swapping [49].

5. Conclusions

NO\textsubscript{x} emission rates from N fertilised soils, manure management practices and forest soils are highly uncertain, because of the lack of good quality datasets. Randomised plot experiments with high spatial and temporal resolution, including a background control across the European climate zones, main crop types and key agricultural management practice are essential to improve modelling of soil NO\textsubscript{x} emissions and develop mitigation options, and thereby improve the predictions of atmospheric O\textsubscript{3} production. This knowledge gap in soil NO\textsubscript{x} emissions could be solved, if long-term monitoring networks, such as ICOS (Integrated Carbon Observation System) and eLTER (i.e. European Long-Term Ecosystem Research Infrastructures), would include soil and canopy NO\textsubscript{x} emissions as mandatory measurements for fertilised croplands, grasslands and forests.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

This work was supported by DEFRA (UK), the GEF-UNEP funded ‘Towards INMS’ project (www.inms.international), and the policy support agreement between Aarhus University and the Danish Government. We wish to thank Dr J Webb for his contribution regarding EEA emission factors. The views expressed in this work are those of the authors and do not represent either DEFRA, GEF or UNEP views.

Ethical statement

There are no ethical concerns regarding this data analysis.

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