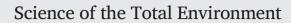
Contents lists available at ScienceDirect







journal homepage: www.elsevier.com/locate/scitotenv

## Predicting field N<sub>2</sub>O emissions from crop residues based on their biochemical composition: A meta-analytical approach



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#### HIGHLIGHTS

- · Meta-analysis to identify best predictors of crop residue N2O emissions
- · Nitrogen returned with residues and residue characteristics were good predictors.
- The predictive power of individual residue characteristics was limited.
- Using correlations between chemical characteristics, we developed a maturity class.
- Immature residues stimulate N<sub>2</sub>O emissions, mature residues have marginal effects.

#### ARTICLE INFO

Article history: Received 7 October 2021 Received in revised form 30 November 2021 Accepted 15 December 2021 Available online 22 December 2021

Editor: Elena Paoletti

Keywords: Biochemical composition Cover crops Crop maturity

\* Corresponding author.

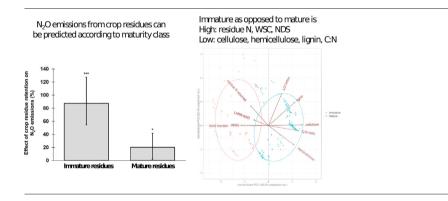
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http://dx.doi.org/10.1016/j.scitotenv.2021.152532

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#### GRAPHICAL ABSTRACT



#### ABSTRACT

Crop residue incorporation is a common practice to increase or restore organic matter stocks in agricultural soils. However, this practice often increases emissions of the powerful greenhouse gas nitrous oxide (N<sub>2</sub>O). Previous metaanalyses have linked various biochemical properties of crop residues to N<sub>2</sub>O emissions, but the relationships between these properties have been overlooked, hampering our ability to predict N<sub>2</sub>O emissions from specific residues. Here we combine comprehensive databases for N2O emissions from crop residues and crop residue biochemical characteristics with a random-meta-forest approach, to develop a predictive framework of crop residue effects on N2O emissions. On average, crop residue incorporation increased soil N2O emissions by 43% compared to residue removal, however crop residues led to both increases and reductions in N2O emissions. Crop residue effects on N2O emissions were best predicted by easily degradable fractions (i.e. water soluble carbon, soluble Van Soest fraction (NDS)), structural fractions and N returned with crop residues. The relationship between these biochemical properties and N2O emissions differed widely in terms of form and direction. However, due to the strong correlations among these properties, we were able to develop a simplified classification for crop residues based on the stage of physiological maturity of the plant at which

Plant species N<sub>2</sub>O emissions the residue was generated. This maturity criteria provided the most robust and yet simple approach to categorize crop residues according to their potential to regulate  $N_2O$  emissions. Immature residues (high water soluble carbon, soluble NDS and total N concentration, low relative cellulose, hemicellulose, lignin fractions, and low C:N ratio) strongly stimulated  $N_2O$  emissions, whereas mature residues with opposite characteristics had marginal effects on  $N_2O$ . The most important crop types belonging to the immature residue group – cover crops, grasslands and vegetables – are important for the delivery of multiple ecosystem services. Thus, these residues should be managed properly to avoid their potentially high  $N_2O$  emissions.

#### 1. Introduction

Crop residue production exceeds five billion tons per year worldwide (Cherubin et al., 2018). A large part of this material is returned to agricultural land as a strategy to restore soil organic matter stocks and build soil fertility. Indeed, together with the return of animal excreta, the incorporation of crop residues is the most important method for farmers to maintain and sustain soil organic matter (Powlson et al., 2008). Although returning crop residues has many benefits for soil health, it may also increase the emission of the powerful greenhouse gas nitrous oxide (N<sub>2</sub>O) (IPCC, 2006; Chen et al., 2016; Hansen et al., 2019). Nitrous oxide emissions from agricultural land account for approximately 60% of the total global anthropogenic N<sub>2</sub>O emissions (Reay et al., 2012), and are therefore considered a key target in the Paris Agreement in order to limit global warming to 1.5 °C or 2 °C (Clark et al., 2020).

Although crop residues generally increase N2O emissions, negative and no effects have also been reported (Xia et al., 2018). The effect of residue retention on N<sub>2</sub>O is difficult to predict since it is mediated via multiple processes. Crop residues might increase N<sub>2</sub>O emissions by providing N for soil microbial processes (Li et al., 2005), including nitrification and also denitrification (Surey et al., 2020). Accordingly, N<sub>2</sub>O emissions from crop residues are often predicted using a fixed emission factor based on the amount of nitrogen (N) returned to the soil with the residues (IPCC, 2019). However, this approach neglects the impact of residue-C addition on microbial dynamics and denitrification processes. Residue C:N ratio, a commonly used index for residue quality, accounts for net N immobilization or mineralization from crop residues, and is also used as predictor of residue-derived N<sub>2</sub>O emissions (Chen et al., 2013). Negative relationships between residue C:N ratio and N<sub>2</sub>O emissions are frequently reported (Chen et al., 2013; Hu et al., 2019), and are generally attributed to decreased N availability due to microbial immobilization of soil mineral N with high C:N residues (Chen et al., 2013). However, this could be an oversimplification, because crop residues differing in their C:N ratio are likely to differ also in terms of soluble and structural contents, which determine the extent and dynamics of residue decomposition and consequently also affect N<sub>2</sub>O production. For example, increases in lignin and polyphenols concentration can be negatively related to N2O emissions (Millar and Baggs, 2004; Garcia-Ruiz and Baggs, 2007), whereas water-extractable organic C may stimulate denitrification (Surey et al., 2020). The direct links between biochemical crop residue characteristics and N<sub>2</sub>O emissions, and particularly the correlations between them, remain poorly understood.

Predicting N<sub>2</sub>O emissions from crop residues is also challenging because these emissions depend on multiple interactions between crop type, environmental conditions, and soil properties (Chen et al., 2013; Lehtinen et al., 2014; Xia et al., 2018; Hu et al., 2019). For example, N<sub>2</sub>O emissions from crop residues are negatively and linearly related to soil clay content (Xia et al., 2018), because high clay content is associated with lower soil porosity and aeration, and thus lower oxygen availability (Skiba and Ball, 2002). However, N<sub>2</sub>O emissions from crop residues are not linearly related to soil pH and SOC, instead peaking at intermediate values of either of those soil properties (Hu et al., 2019). Residue return stimulates N<sub>2</sub>O emissions more strongly in regions with a warmer climate (e.g., warm temperate and subtropical) compared to cool temperate zones, probably due to increased decomposition rate. Residue-derived N<sub>2</sub>O emissions may be negatively associated with mean annual precipitation (Xia et al., 2018); yet, other studies showed increased emissions at intermediate soil water content (i.e. water filled pore space 60–90%; Chen et al., 2013), perhaps due to confounding effects via soil type. While previous meta-analyses have evaluated these factors individually, their hierarchical importance has been overlooked.

The objective of this meta-analysis was to synthesise the results of field studies measuring the effect of crop residue retention on soil N<sub>2</sub>O emissions. We aimed to identify the main drivers of soil N<sub>2</sub>O emissions associated with the application of crop residues and to rank their importance. Our study differs from previous meta-analyses in several aspects: we only collected information from field studies to avoid unrealistic emissions from crop residues; used the most comprehensive approach so far to gather biochemical characteristics of crop residues; used a random-meta-forest approach, which accounts for multiple drivers simultaneously including non-linear relationships; and developed and tested for the first time a simplified classification based on the stage of residue maturity. We hypothesized that the relationships between crop residue biochemical characteristics and N<sub>2</sub>O emissions can be unfolded to develop a predictive framework of crop residue effects on N<sub>2</sub>O emissions.

### 2. Materials and methods

#### 2.1. Data collection and data gap filling

#### 2.1.1. Crop residue and soil N<sub>2</sub>O emissions database

We collected data on N<sub>2</sub>O emissions from field experiments with crop residue retention treatments across the world. We defined crop residues as any constituent of a crop (above- and below-ground) returned to the soil. These may originate from cash crops, cover crops, green manures, grassland or others. Depending on the crop, the residues may include below-ground and above-ground biomass, except for root crops for which it refers to above-ground biomass only since most of the roots are harvested. The crop residues treated through biogas fermentation, composting and other methods were not included. Another difference with respect to previous meta-analyses is that we included grassland termination data in our database, because temporary grasslands make up a substantial share of arable land in Europe, especially in the Scandinavian countries and in some regions with high livestock densities, e.g. the Netherlands, Belgium and Bretagne, France (Lesschen et al., 2014). Peer-reviewed research articles published before 1 January 2021 were searched in Google Scholar and Web of Science with the following keywords: "residue" OR "straw" OR "stubble" AND "nitrous oxide" OR "N2O" OR "greenhouse gas" OR "GHG" OR "emission factor". The initial search resulted in 337 publications. We also included results from two unpublished studies conducted by authors of this meta-analysis (Bleken et al., n.d.; Ernfors et al., n.d.). All publications were screened and included in the database if they met the following criteria: (i) the experimental design had to be sufficiently detailed regarding the treatments and recent history; (ii) included treatment replicates; and (iii) were conducted under realistic field conditions. All the studies included pairwise comparisons in which all factors (e.g., fertilization, irrigation, tillage practices) were the same for the treatment (with crop residue retention) and control (with crop residue removal) groups. For grassland, we compared permanent grassland (control) to grassland termination (treatment), the analogous practice to crop residue incorporation in these systems. Note that the control treatments do not always mean residue-free soil and its definition varies among studies, e.g. in green manure studies, the control may be a treatment without the

addition of herbage with the presence of roots or a fallow plot where the roots were removed. The selected studies (n = 78) provided 367 pairwise comparisons. Cumulative N<sub>2</sub>O emissions (g N<sub>2</sub>O-N ha<sup>-1</sup>) for each crop residue treatment and control were extracted together with a measure of variance and the number of replicates. The length of the studies ranged from 5 to 575 days. To account for potential biases due to the inclusion of short-term studies (<40 days), we ran an initial sensitivity analysis excluding short-term studies and it did not affect our main results. For each study in our dataset, we tabulated details on the site and experimental conditions. When data were only presented graphically, WebPlotDigitizer was used to extract data points. If relevant information was not reported, the authors were contacted to supply the missing information.

#### 2.1.2. Data aggregation

We identified 21 factors as potential drivers of N<sub>2</sub>O emissions from crop residues (Table 1). These factors related to crop type, residue type, residue properties, soil properties, and climatic conditions, and included both categorical and continuous factors. A detailed data overview is presented in a publicly available database accompanying this publication (Rittl et al., n. d.). For crop type, we distinguished between cereal (mature harvested cereals and rapeseed), rice, cover crop (including immature cereals, grasses, clovers, Brassica spp. cultivated as cover crops or one-year green manures), grassland, grain legumes, vegetables, and sugar cane, and double cropping (combination of two species at different biomass ratios). For soil properties, soil texture was divided into clay (clay, sandy clay, silt clay), loam (loam, clay loam, silty clay loam, silt loam, sandy loam, sandy clay loam, fine silt, silt), and sand (sand, loamy sand, fine sand) (Quemada et al., 2013). Soil pH values measured in CaCl<sub>2</sub> were converted to H<sub>2</sub>O-based values using the equation proposed by Ros et al. (2020). Soil pH values in our dataset ranged from 4.3 to 9.3, and were grouped into acidic (<6), neutral (6–7), and alkaline (>7) soils. We converted organic matter content to organic C content using the standard conversion factor of 0.58 (Pribyl, 2010).

We calculated the Aridity Index (AI) as the ratio of mean annual precipitation to mean annual potential evapotranspiration (UNEP). The long-term mean annual precipitation was extracted from the publications, and the long-term mean annual potential evapotranspiration for each site was extracted from the Global Aridity Index geodatabase (https://cgiarcsi.community/data/global-aridity-and-pet-database/). We used the classification of wet (AI > 1) and dry land (AI < 1) utilized by IPCC (2019) to differentiate N<sub>2</sub>O emission factors from N sources (including organic amendments such as crop residues).

We assessed the impact of study duration on a thermal time basis, using the normalized time approach proposed by Andren and Paustian (1987), and applied successfully to soil C and N mineralization, gross N fluxes and C-N models (Aita et al., 1997; Brisson et al., 2009). In short, the total duration of the experiment was converted to number of days at a reference temperature of 15  $^{\circ}$ C (i.e. ND15), following the approach described by Thiébeau and Recous (2017).

#### 2.1.3. Crop residue quality characteristics and categorical groups

From each study in our dataset, we extracted information on the chemical characteristics of crop residues. We calculated the N returned with crop residues from the residue N concentration and the dry matter (DM) added. When C and N added were reported, we calculated crop residue C:N ratio. When these data were not provided, mean values were taken from the crop residue quality dataset provided by Thiébeau et al. (2021) for the same species or group of species. Residue concentration of soluble fraction (neutral detergent soluble, referred to as soluble NDS fraction), hemicellulose, cellulose, and lignin (according to Van Soest (1963)) were tabulated when these were reported. When these data were not available, which was the case for the vast majority of studies included in the database (>90%), we used crop residue quality data from Thiébeau et al. (2021). Previous research has shown that, on a global scale, most of the variation in plant characteristics is represented by species identity (Kattge et al., 2011). Accordingly, the use of a global dataset for residue biochemical characteristics was deemed appropriate for our global meta-analysis. Hemicellulose, cellulose and lignin relative contents (% total DM), indicative of the composition of the insoluble residue fraction (i.e. the plant cell walls), were used, the difference between their sum and 100%, being the soluble NDS fraction. The lignocellulose index (LCI) [lignin: (lignin + cellulose + hemicellulose)] was used as a criteria to express the recalcitrance of the plant cell wall (Herman et al., 2008). Water soluble C (WSC) determined after water extraction, was expressed as a percentage of total C. Crop C:N ratio was divided into three categories (<20, 20-60, >60).

We used two criteria to categorize crop residues: according to maturity stage, and to the type of residue generated. Crop residues were classified as mature or immature, based on the stage of physiological maturity of the plant at which the residue were generated (Sylvester-Bradley et al., 2015). This could be by cultivation practice (harvesting of annual crops at the end of the cycle, harvesting of root or vegetable crops, mechanical destruction, grassland mowing or grassland renewal) or possibly naturally (senescence), irrespective of the plant type. Incorporation of immature residues into the soil happens during: (i) destruction of green cover crops or

#### Table 1

Overview of factors used to categorize the crop residues included in our analysis.

Groups	Factors	Number of observations	Study categories and range for continuous factors
Residue-N	N returned with crop residues (kg N $ha^{-1}$ yr <sup>-1</sup> )	297 <sup>a</sup>	5 to 418
Crop type	Crop type	367	Cereal, cover crop, grassland, grain legumes, rice, sugar cane, vegetable, double cropping
Residue type	Type of residue generated	367	Green plant biomass, mature above ground biomass, senescent plant biomass, straw
	Maturity index	367	Immature, mature
Residue quality	Residue C:N ratio	357 <sup>a</sup>	<20, 20–60, >60
	Soluble NDS (% total DM)	344 <sup>a</sup>	6 to 71
	Cellulose (% total DM)	346 <sup>a</sup>	10 to 49
	Hemicellulose (% total DM)	345 <sup>a</sup>	6 to 55
	Lignin (% total DM)	352 <sup>a</sup>	2 to 26
	Lignocellulose Index (LCI)	344 <sup>a</sup>	0.041 to 0.306
	Water soluble carbon (% total C)	288 <sup>a</sup>	3 to 68
Soil properties	Soil texture	222	Clay, loam, sandy
	Clay (%)	219	3.1 to 71%
	Soil pH	288 <sup>a</sup>	Acidic (<6), neutral (6–7), alkaline (>7)
	Soil organic carbon (g C kg <sup>-1</sup> SDW)	310 <sup>a</sup>	2 to 98
	Soil bulk density (g $\text{cm}^{-3}$ )	178	0.76 to 1.7
	Soil total N (g N kg <sup>-1</sup> SDW)	262	0.14 to 6.4
Climatic conditions	Annual mean precipitation (mm)	367 <sup>a</sup>	350 to 2115
	Annual mean temperature (°C)	227	5.3 to 27.4
	Aridity index	361 <sup>a</sup>	<1,>1
	Normalized days at 15 °C	192	4.7–1001

<sup>a</sup> Factors included as potential predictors in the random-meta-forest approach.

catch crops; (ii) pre-harvest destruction of the green haulm of potatoes, carrots, sugar beet and other root crops; (iii) harvest of brassicas and other vegetable crops (e.g., cauliflowers, cabbages and green salads), where leafy material is left in the field; (iv) ploughing down of herbage of grass leys (Sylvester-Bradley et al., 2015). In our database, the classification of immature residues refers to cover crops, vegetable residues and grassland renewal. Residues were classified as mature for a range of post-crop harvest residues such as straw and stem residues from cereals, rice, grain legumes, oilseed, sugar cane and others. Second, we categorized studies according to the type of residue generated (Table 1). Residues types were grouped into mature above-ground biomass, straw, green plant biomass, and senescent plant biomass (Table 1). Mature above-ground biomass encompassed above-ground residues at maturity for all crops except cereals. Straw indicates above-ground residues of cereals including rice at maturity. Green plant biomass is the total plant harvested at vegetative stage (i.e. cover crops, grassland renewal, vegetables). Senescent plant biomass refers to leaves of perennial crops, in our case only sugar cane.

#### 2.1.4. Dataset syntheses

The database included studies from 69 sites in 20 countries. Europe contributed 41% of the comparisons (n = 148). For Europe, 32% of the data were from residues classified as cover crop; 80% as green plant biomass; 60% of the N<sub>2</sub>O comparisons were from experiments where fertilizer was applied to either the growing crop generating the residue, or at some point following residue return. Regarding climate, 63% of our comparisons originated from field sites in dry climates (AI <1), and 37% from sites in wet climates (AI >1). The range of each continuous factor is shown in Table 1.

#### 2.2. Data analysis

We used the log response ratio (LnRR) as effect size, which is a common metric in meta-analyses (Hedges et al., 1999; Osenberg et al., 1999). We performed a weighted mixed-effects meta-analysis, using the rma.mv function of the 'metafor' package (Viechtbauer, 2010), including "study" as a random effect because several studies contributed more than one effect size. Effect sizes from individual comparisons were weighted by the inverse of their variance. Missing variances were estimated using the average coefficient of variation across the dataset (van Groenigen et al., 2017). We used a Wald test to evaluate statistical differences between subgroups within categories at p-value < 0.05.

We used a random-meta-forest approach to identify the most important predictors of crop residue effects on N2O emissions. This approach is based on the machine learning random forest algorithm. This approach incorporates the variance and weight of each study as in classic meta-analysis, is robust to overfitting, allows for numerous predictors, and accounts for nonlinear relationships (Terrer et al., 2019, 2021). Among the 21 potential factors, we discarded those based on expert knowledge to avoid possible artifacts associated with arbitrary category definitions (i.e. crop type, residue type, maturity class) (Table 1), and those with more than 30% missing data (soil texture, clay, annual temperature, soil bulk density, soil total N, and ND15). The final dataset consisted of 267 observations with information on 12 predictors, which are indicated in Table 1. Using 'metaforest' (Van Lissa, 2017), we performed variable pre-selection with 10,000 iterations and 100 replications, using a recursive algorithm in the preselect function from metafor. Predictors with negative variable importance were dropped using the preselect\_vars function. Predictors with positive predictive performance were then used to optimize the model. Parameters of the meta-forest model were optimized using the train function from the caret package (Kuhn, 2008), and calculated 10-fold cross validated R<sup>2</sup> with 75% of the data used as training data and 25% as validation data. We generated partial dependence plots to depict the association of each selected predictor with the effect size, while accounting for the average effect of all other predictors.

To check the robustness of our random-meta-forest approach, we ran another mixed-effects meta-regression model selection procedure with the six most important predictors identified by random forest analysis. The model selection was based on maximum likelihood estimation, which was carried out with the 'glmulti' package in R (Calcagno and de Mazancourt, 2010). The importance of each predictor was computed as the sum of Akaike weights for models that included this predictor. A cutoff of 0.8 was set to differentiate between essential and non-essential predictors (Chen et al., 2020; Terrer et al., 2019).

Nitrogen fertilizer application is one of the most important drivers of  $N_2O$  emissions, and it may affect the relationships between our identified controlling factors and  $N_2O$  emissions from crop residues, as observed for organic amendments (Charles et al., 2017) and for crop residues (Shan and Yan, 2013). Our initial analyses confirmed the importance of N fertilizer addition (p < 0.01). Thus, we used 'metafor' to test for interactions between fertilizer application (i.e. with or without application) and each of the most important predictors identified by the random-meta-forest approach. When the interaction term was non-significant, we proceeded with the analyses using the complete dataset; when the interaction was significant, we conducted separate analyses for studies with and without fertilizer application. We fitted linear and quadratic meta-regressions to identify the best model describing the relationships between the most important predictors and LnRR, based on the Akaike information criterion (AIC); the model with lower AIC was retained.

Principal component analyses (PCA) were performed with R (prcomp available in the base package, and the 'ggbiplot' package; Vu, 2011) to visualize the main biochemical characteristics of mature and immature residues, and of the main crop type.

#### 3. Results

#### 3.1. Overall effect of crop residues on N<sub>2</sub>O emissions

Averaged across all studies, crop residue retention increased N<sub>2</sub>O emissions by 43% (95% CI: 23–66%; p < 0.001). However, the differences between crop types were large (Fig. 1). Crop residues increased N<sub>2</sub>O emissions for cover crops (50%, CI: 19–88%), grasslands (216%, CI: 114–366%) and vegetables (183%, CI: 85–334%), but not for cereals, grain legumes, rice or sugar cane (Fig. 1).

#### 3.2. Main predictors of crop residue induced N<sub>2</sub>O emissions

Our random-meta-forest approach identified N returned with crop residues, WSC, cellulose, lignin, hemicellulose and soluble NDS fraction as the most important predictors of crop residue effects on  $N_2O$  emissions (Supp. Fig. 1). Model selection analysis based on Akaike weights confirmed the importance of these predictors, as all of them reached the threshold of 0.8 (Supp. Fig. 2). The importance of residue quality properties was therefore higher than that of the soil properties and climatic factors included in the random-meta-forest analysis (Table 1).

The amount of N returned (kg N ha<sup>-1</sup> yr<sup>-1</sup>) with the crop residues affected the N<sub>2</sub>O emissions (AIC = 481.49, n = 298), and the overall effect of the amount of residue-N was not modified by the application of fertilizer N (p > 0.05). Nitrous oxide emissions from crop residues increased with the amount of residue-N returned, reaching a maximum at ~200–250 kg N ha<sup>-1</sup> yr<sup>-1</sup> applied (Fig. 2). Crop residue-derived N<sub>2</sub>O emissions decreased with increasing values of cellulose, whereas intermediate hemicellulose values (~30%) were associated with the lowest LnRR (Fig. 2). The effects of cellulose (AIC = 592.87, n = 346) and hemicellulose (AIC = 612.05, n = 345) on N<sub>2</sub>O emissions from crop residues were not affected by fertilizer application either.

Nitrogen fertilizer application affected the relationship between crop residue N<sub>2</sub>O emissions and WSC (with fertilizer: AIC = 208.02, n = 181; without fertilizer: AIC = 133.58, n = 70), soluble NDS fraction (with: AIC = 297.59, n = 210; without: AIC = 148.13, n = 79), and lignin (with: AIC = 313.18, n = 213; without: AIC = 153.08, n = 84), but the trends were in general consistent with and without fertilization (Fig. 3). N<sub>2</sub>O emissions from crop residues increased with WSC values and when the residue soluble NDS fraction exceeded 40%. High lignin content in

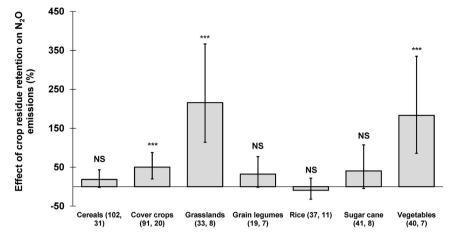


Fig. 1. Relative increase in field N<sub>2</sub>O emission (mean ± 95% CIs) with crop residue retention, as affected by crop type. The number of observations and studies are shown in parentheses. See section 2.1.2. for detailed descriptions of crop categories.

the crop residues corresponded with smaller treatment effects on  $\rm N_2O$  emissions for studies without added fertilizer, but not for studies with added fertilizer.

#### 3.3. Residue type and $N_2O$ emission

Maturity class had a strong influence on the magnitude of N<sub>2</sub>O emissions (p < 0.001), with greater emissions from immature than from mature residues (Wald-type test, p < 0.001) (Fig. 4). The type of residue significantly affected crop residue derived N<sub>2</sub>O emissions (p < 0.001). Green plant biomass increased N<sub>2</sub>O emissions by 89%, and mature aboveground biomass by 40% (Supp. Fig. 3). Conversely, straw and senescent plant biomass did not increase N<sub>2</sub>O emissions.

#### 3.4. Predictors with lower importance

The response of N<sub>2</sub>O emissions to crop residue incorporation was not explained by the lignocellulose index (LCI), annual mean precipitation (i.e. these two predictors were not selected by the random-meta-forest approach), soil texture, soil bulk density, annual mean temperature or study length either expressed by calendar day or by standard day (ND15) (i.e. these predictors had no significant effect on LnRR based on mixed-effects meta-analysis). However, simple linear meta-regressions indicated that residue C:N ratio (p < 0.001), soil pH (p = 0.014), SOC (p = 0.011), soil N (p = 0.002) and Aridity Index (p = 0.021) exerted a small but significant influence on crop residue N<sub>2</sub>O emissions (Supp. Figs. 4, 5). High C:N ratios (above c. 60), neutral soil pH, low SOC and low soil N content led to lower N<sub>2</sub>O emissions. The effect size of applying crop residues in humid climates was 2 times larger than in dry climates.

#### 3.5. Relationship between residue biochemical characteristics

A PCA analysis was used to determine the association between residue biochemical characteristics, grouped for maturity class and crop type (Fig. 5). The first two principal components accounted for 72.4% of the total variance. We found strong relationships between residue biochemical characteristics: WSC, NDS fraction and residue N returned were positively correlated among them, and negatively related to cellulose, hemicellulose and C:N ratio. Immature residues were characterized by high WSC, NDS fraction and N returned with low cellulose, hemicellulose, lignin fractions, LCI and C:N ratio; the opposite was true for mature residues (Fig. 5a). Cover crop residues generally contained a high soluble NDS fraction and WSC, vegetable residues returned relatively large amounts of N, and grassland residues presented low lignin concentration (Fig. 5b). Cereals and sugar cane residues were characterized by a high cellulose and hemicellulose content, and a high C:N ratio.

#### 4. Discussion

#### 4.1. Magnitude of N<sub>2</sub>O emissions as affected by crop type

In line with other meta-analyses (Chen et al., 2013; Hu et al., 2019; Muhammad et al., 2019; Shan and Yan, 2013), we found a significant stimulation of soil N<sub>2</sub>O emission following the application of residues from vegetables, cover-crops and grasslands renewal, but not from cereals, grain legumes, rice and sugar cane. For vegetables, we observed an average increase in N<sub>2</sub>O emission of around 183%, which is higher than reported in the meta-analysis of Shan and Yan (2013) for lettuce and bean residues (123% and 138% increase, respectively). Our finding that incorporation of cover crop residue stimulated N<sub>2</sub>O emissions is consistent with Muhammad et al. (2019), although they only found increased N<sub>2</sub>O emissions for legume-based cover crops. As we did not distinguish between residues from cover crops with and without legumes, we cannot directly compare our results with theirs.

#### 4.2. The main drivers behind crop residue N<sub>2</sub>O emissions

Residue biochemical characteristics were strong predictors of N<sub>2</sub>O emissions from crop residues. Easily degradable fractions (WSC, soluble NDS fraction), structural fractions (cellulose, hemicellulose and lignin), and N returned with crop residues (resulting from residue N concentration and biomass) had the most predictive power. Crop residues rich in easily degradable fractions increased N<sub>2</sub>O emissions, probably by providing C sources for biological N transformation processes and by accelerating microbial growth (Sahrawat and Keeney, 1986). The easily degradable fractions can also promote N<sub>2</sub>O emissions by creating an anaerobic environment, and by providing energy sources for nitrifiers, and electron acceptors for bacterial and fungal denitrifiers (Palatinszky et al., 2015; Surey et al., 2020). Indeed, during short-term anoxic conditions due to high O2 demand from microbial activity, denitrifying organisms rely on soluble compounds from fresh residues as major C sources (Surey et al., 2020). Conversely, structural fractions, which are relatively resistant to decomposition, were negatively related to N2O emissions from crop residues. This is consistent with a large body of literature showing that recalcitrant materials such as stubbles and roots can take longer to mineralize, and can lead to net immobilization of soil mineral N (e.g. Kuzyakov, 1999; Gentile et al., 2008). The stimulatory effect of N returned with crop residues on N2O emissions is well established (Li et al., 2016; Hansen et al., 2019), as the release of residue N after mineralization increases the availability of soil mineral N for nitrification and denitrification, which are likely to be source processes under different environmental conditions.

The divergent relationships between individual biochemical characteristics and  $N_2O$  emissions makes predicting  $N_2O$  emissions from crop

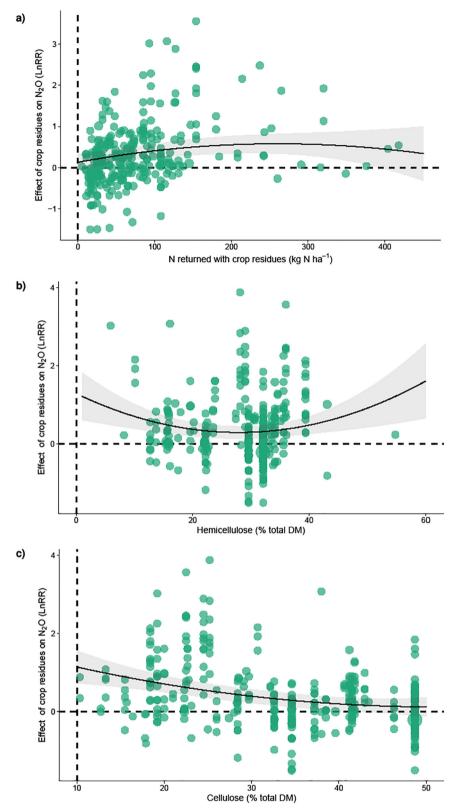
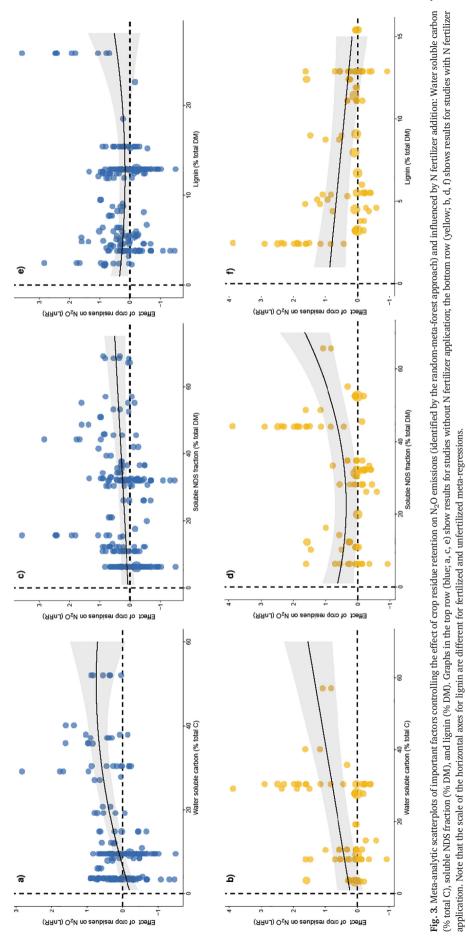


Fig. 2. Meta-analytic scatterplots of important factors controlling the effect of crop residue retention on  $N_2O$  emissions (identified by the random-meta-forest approach); a) N returned with crop residues (kg N ha<sup>-1</sup> yr<sup>-1</sup>), b) residue hemicellulose concentration (% DM), and c) residue cellulose concentration (% DM).

residues challenging. However, our results shed light on how to overcome these difficulties. This is because crop residue biochemical characteristics are correlated, as shown by the multivariate analysis (Fig. 5). From a quantitative point of view, this is of course linked to the characterization used (i.e. Van Soest method), with values representing relative proportions in fractions of increasing recalcitrance, expressed as a function of the total mass (Soest, 1963; Soest and Wine, 1967). The biological explanation is that the distribution of the different pools within the plant tissues changes with plant development, with characteristic compositions linked to given developmental stages and functions (e.g. leaf vs. stem vs. root). For



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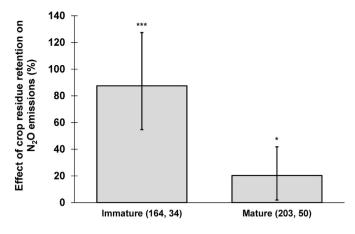


Fig. 4. Relative increase in field  $N_2O$  emission (mean  $\pm$  95% CIs) with crop residue incorporation, as affected by residue maturity class. The number of observations and studies are shown in parentheses. See section 2.1.3. for a detailed description of the maturity criteria.

example, residues from cereals harvested at mature stages have less soluble and more structural fractions than cereals harvested earlier in the growth cycle (Bertrand et al., 2009), causing a negative association between soluble and structural fractions (Supp. Fig. 6). Our results also show a strong positive correlation between residue N returned and the soluble NDS fraction, confirming previous work (Schmatz et al., 2020). These biochemical associations indicate that crop residues can be categorized beyond single residue properties (e.g. N content, cellulose, hemicellulose, lignin), and that aggregate indicators of residue composition are probably the most useful predictors of residue effects on soil  $N_2O$  emissions.

# 4.3. Maturity class: a new and robust predictor of $N_2O$ emissions from crop residues

We tested a simple aggregate indicator based on the stage of maturity to classify crop residues with respect to their potential to induce N<sub>2</sub>O emissions. The maturity stage, used to categorize crop residues as mature and immature residues, showed a large impact on soil N<sub>2</sub>O emissions. Immature residues, with an overall composition of high content of WSC, soluble NDS

fraction and N returned, and low cellulose, hemicellulose, lignin, LCI and C: N ratio, stimulated the effect of residues on N2O emissions. Immature residues were composed by cover crops, vegetable residues and grassland renewal. Incorporation of mature residues on the other hand, had a weak influence on N<sub>2</sub>O emissions. Mature residues included a wide range of cereal residues in the form of straw. This simple and yet robust categorization offers a tool for predicting N<sub>2</sub>O emissions from crop residues obtained under a broad range of pedoclimatic conditions, encompassing intrinsically the different crop properties and management practices associated with their cultivation. Previous syntheses with a similar focus advocated for the use of residue C:N ratio as the best predictor of N<sub>2</sub>O emissions from crop residues (Chen et al., 2013; Hu et al., 2019; Muhammad et al., 2019; Shan and Yan, 2013). Yet, our random-meta-forest approach revealed that the C:N ratio was not among the most solid predictors of residuederived N<sub>2</sub>O emissions (Supp. Fig. 1). This may be because C:N ratio only accounts for the balance between residue N mineralization/immobilization, whereas maturity class, by integrating a wide range of biochemical properties, also addresses other controlling factors of N<sub>2</sub>O emissions such as C availability and soil O<sub>2</sub> consumption. Therefore, the use of our simple classification according to maturity class provides new opportunities to identify crop residues requiring targeted agronomic practices for reducing N<sub>2</sub>O emissions, inform agricultural policy measures designed to mitigate climate change, and predict the potential impacts of crop rotations on N<sub>2</sub>O emissions.

#### 4.4. Impact of abiotic conditions

Our random-meta-forest approach indicated that soil texture, soil bulk density, mean annual precipitation, mean annual temperature, and study length had little predictive power regarding  $N_2O$  emissions from crop residues. Our hierarchical approach, using only data from field experiments that are representative for on-farm conditions, indicates that although some of these factors exert an influence on crop residue decomposition and associated  $N_2O$  emissions (e.g. mean annual precipitation and soil texture; Chen et al., 2013; Xia et al., 2018; Hu et al., 2019), their overall importance is much lower than that of residue biochemical properties, and also lower than that of other abiotic conditions.

The most relevant soil and environmental factors among the ones tested in our study were soil pH, SOC, soil N and Aridity Index, with neutral soil pH, low SOC and low soil N content leading to lower N<sub>2</sub>O emissions. For

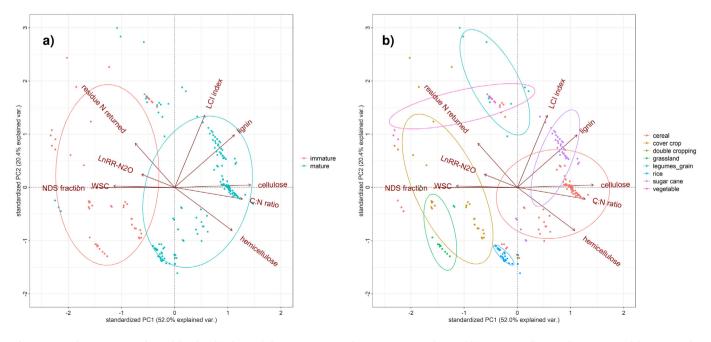


Fig. 5. Principal component analysis of the plant biochemical characteristics according to a) maturity class, and b) crop type. Ellipses indicate 95% confidence intervals.

the soil properties, these results are broadly in agreement with previous meta-analyses (Chen et al., 2013; Xia et al., 2018; Hu et al., 2019). The impact of SOC and soil N reflects their role as C and N sources for N2Oproducing microorganisms; low soil pH (acidic) increases N<sub>2</sub>O fluxes due to impairment of periplasmatic N<sub>2</sub>O reductase (Bergaust et al., 2010; Bakken et al., 2012). While the importance of soil parameters has been summarized in previous meta-analyses, our study is the first one showing the role of the Aridity Index for crop residue-derived N2O emissions. The effect size of applying crop residues in humid climates was 2 times greater than in dry climates. As posited by Rochette et al. (2008), using annual precipitation can lead to biased conclusions because sites with precipitation exceeding 1000 mm are usually only found under humid climates, which could be tropical or temperate. The ratio between precipitation and evapotranspiration corrects for this bias, which explains why the Aridity Index was a better predictor of N<sub>2</sub>O emissions from crop residues than annual precipitation and annual temperature alone, and therefore its use should be promoted.

#### 4.5. Implications

The strong positive effect of immature crop residues on N<sub>2</sub>O emissions has important implications for current efforts to achieve sustainable cropping systems, as residues falling in this category originate from cover crops, grasslands, and vegetables. Cover crops provide a wide range of ecosystem services (Haruna and Nkongolo, 2015), including reductions of N losses in the form of nitrate leaching (Abdalla et al., 2019). However, the increase in N<sub>2</sub>O emissions after their incorporation may partly compromise these benefits. Our analysis only included cover crop effects on N<sub>2</sub>O emissions after incorporation, but it is possible that cover crops may reduce soil N2O emissions during their growing phase by reducing soil mineral N availability, thereby countering their afterlife effects (Han et al., 2017). Including temporary grasslands in rotation with annual crops may reduce water pollution (Parish et al., 2012), soil erosion (Pimentel et al., 2012), and pests (Werling et al., 2014), and promote biodiversity (Meehan et al., 2010) and soil C sequestration (Beniston et al., 2014). Yet, our results suggest that the increased N<sub>2</sub>O emissions after grassland termination may largely offset the GHG balance benefits of increased soil C storage. Regarding crop residues of vegetables, it is worth noting that global vegetable production is rapidly expanding due to health guidelines advocating for an increase in vegetable consumption (Norris and Congreves, 2018). However, the high N<sub>2</sub>O emissions induced by their residues (and during the vegetation period; Qasim et al., 2021), and the high requirements for fertilizers, irrigation, and tillage needed for vegetable production call for an increased research effort to improve the environmental sustainability of these systems.

Our findings also have major implications for calculating N<sub>2</sub>O emission factors from crop residues. First, the relationship between supplied N with crop residues and N<sub>2</sub>O emissions is not linear (Fig. 2), which challenges the use of an emission factor that assumes linearity between N returned in residues and emitted N<sub>2</sub>O as assumed by the IPCC Tier 1 approach (IPCC, 2019). Second, using one single factor as residue N is not appropriate to explain the magnitude of the response of N<sub>2</sub>O emissions to the application of crop residues across multiple sites, years and cropping systems. The use of integrative quality criteria such as maturity class or residue type, are more promising approaches to constrain the variability regarding N<sub>2</sub>O emissions from crop residues. This classification could therefore be used to improve the estimation of N<sub>2</sub>O emission factors from crop residues, and it may be easily incorporated in the IPCC guidelines to estimate N<sub>2</sub>O emissions from agricultural sources.

#### CRediT authorship contribution statement

<u>Diego Abalos</u>: conceptualization, formal analysis, writing - review and editing; <u>Tatiana Rittl</u>: data curation, writing - original draft, review and editing; <u>Sylvie Recous</u>: conceptualization, data curation, writing - review and editing; Pascal Thiébeau: investigation, resources, data curation;

Cairistiona F. E. Topp: data curation, review and editing; Kees Jan van Groenigen: formal analysis, review and editing; Klaus Butterbach-Bahl: review and editing; Rachel E. Thorman: data curation, review and editing; Kate E. Smith: review and editing; Ishita Ahuja: data curation, review and editing; Jørgen E. Olesen: project management, review and editing; Marina A. Bleken: review and editing; Robert M. Rees: review and editing; Sissel Hansen: conceptualization, coordination, writing - review and editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgments

This study was supported by the FACCE ERA-GAS Cofund (European Research Area - for mitigation of Greenhouse gases from Agri- and Silviculture; id number: 696356), project ResidueGas (https://projects.au.dk/ residuegas/). The Danish funding was received from Innovation Fund Denmark (project 7108-00005B). The French funding was received from the Agence Nationale de la Recherche (ANR-17-EGAS-0003) and the National Institute for Food, Agriculture and the Environment (INRAE). The authors acknowledge funding in the UK from the Department for Environment, Food and Rural Affairs (Defra) and the Scottish Government's Strategic Research Programme. The authors acknowledge funding in Norway from the Research Council of Norway (Project No.: 276389). We would like to thank the researchers who were contacted and kindly supplied any missing information: Liz Baggs, Søren O. Petersen, Claudia Wagner-Riddle, Yuefang Zhang, Pierre-André Jacinthe, Caroline Buchen, Kari Dunfield, Maria Ernfors, Katharina Kesenheimer, Xiaoxi Li, Marco Siqueira Neto, Thorsten Reinsch, Reiner Ruser, Reinhard Well, Khagendra Baral, and Jaber Rahimi. The Chamber of Agriculture Lower-Saxony contributed with climatic data. Special thanks to Christian Werner for extraction of geodata.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2021.152532.

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