# <sup>1</sup> Measurement of N<sub>2</sub>O emissions over the whole year is

## 2 necessary for estimating reliable emission factors

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#### 23 Abstract

Nitrous oxide emission factors (N<sub>2</sub>O-EF, percentage of N<sub>2</sub>O-N emissions arising from applied 24 25 fertilizer N) for cropland emission inventories can vary with agricultural management, soil properties and climate conditions. Establishing a regionally-specific EF usually requires the 26 measurement of a whole year of N<sub>2</sub>O emissions, whereas most studies measure N<sub>2</sub>O emissions 27 28 only during the crop growing season, neglecting emissions during non-growing periods. However, the difference in N<sub>2</sub>O-EF ( $\Delta$ EF) estimated using measurements over a whole year 29 (EF<sub>wy</sub>) and those based on measurement only during the crop-growing season (EF<sub>gs</sub>) has 30 received little attention. Here, we selected 21 studies including both the whole-year and 31 growing-season N<sub>2</sub>O emissions under control and fertilizer treatments, to obtain 123 ΔEFs from 32 33 various agroecosystems globally. Using these data, we conducted a meta-analysis of the  $\Delta EFs$ by bootstrapping resampling to assess the magnitude of differences in response to 34 35 management-related and environmental factors. The results revealed that, as expected, the  $EF_{wy}$ was significantly greater than the  $EF_{gs}$  for most crop types. Vegetables showed the largest  $\Delta EF$ 36 (0.19%) among all crops (0.07%), followed by paddy rice (0.11%). A higher  $\Delta EF$  was also 37 identified in areas with rainfall  $\geq$  600 mm yr<sup>-1</sup>, soil with organic carbon  $\geq$  1.3% and acidic soils. 38 Moreover, fertilizer type, residue management, irrigation regime and duration of the non-39 growing season were other crucial factors controlling the magnitude of the  $\Delta EFs$ . We also 40 found that neglecting emissions from the non-growing season may underestimate the N2O-EF 41 by 30% for paddy fields, almost three times that for non-vegetable upland crops. This study 42 highlights the importance of the inclusion of the non-growing season in the measurements of 43 N<sub>2</sub>O fluxes, the compilation of national inventories and the design of mitigation strategies. 44

### 46 Capsule

47	Fallow-season $N_2O$ emissions must be included when calculating emission factors (EFs);
48	neglecting them lowers the EFs by 30% for paddy rice and 10% for non-vegetable crops.
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50 Key words: nitrous oxide, greenhouse gas, fallow, residual fertilizer N, nitrogen use efficiency

#### 51 **1 Introduction**

Nitrous oxide (N<sub>2</sub>O) is a long-lived greenhouse gas (GHG) with a mean atmospheric lifetime 52 53 of ~120 years and has a global warming potential (GWP) of ~300 times that of CO<sub>2</sub> over a 100year period (Myhre et al., 2014). N<sub>2</sub>O is the most significant ozone-depleting substance and is 54 predicted to remain the largest during the 21st century (Ravishankara et al., 2009). About 50% 55 of global anthropogenic N<sub>2</sub>O emissions are from agricultural soils, and the percentage has been 56 57 rising since the 1950s, due to the widespread application of synthetic nitrogen (N) fertilizers (Myhre et al., 2014; Shang et al., 2019). Microbial nitrification and denitrification in managed 58 59 and natural soils contribute approximately 70% of globalN2O emissions (Braker and Conrad, 2011). Previous studies have shown that these two processes are regulated mainly by available 60 N pools (synthetic or organic), soil characteristics and environmental conditions (e.g., soil 61 temperature, water content, bulk density and pH) (Abdalla et al., 2009; Butterbach-Bahl et al., 62 2013; Zhou et al., 2015). An inventory framework was devised by the Intergovernmental Panel 63 on Climate Change (IPCC) to quantify the impacts of N availability and other environmental 64 factors on N<sub>2</sub>O emissions from agricultural soils using a N<sub>2</sub>O emission factor (N<sub>2</sub>O-EF) 65 approach at regional scale (IPCC, 2000, 2006). 66

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Emission factors (EFs) are a pragmatic approach, widely used for the compilation of national 68 GHG inventories. The N<sub>2</sub>O-EF is the percentage of N<sub>2</sub>O-N emissions arising from applied N 69 from synthetic and organic fertilizers. The IPCC Tier 1 approach uses a global default EFs, but 70 at Tier 2 these default values can be replaced by country-specific EFs, based on local 71 72 measurements covering an entire year, including growing and non-growing (fallow) seasons, reflecting the varying impacts of environment and management over time (IPCC, 2000, 2006; 73 Wang et al., 2019). The non-growing period, or fallow season, is a period of the year when no 74 crop is growing in arable lands and occurs between crop harvest and the sowing or transplanting 75

of the following crop. Since there can be multiple cropping seasons within a year, e.g., rice-76 wheat rotation during a year in the Chinese Taihu Lake region (Zhao et al., 2012), the non-77 78 growing season is the sum of the multiple fallow periods for each crop of the rotation. However, 79 emission measurements from most field studies, from which the EFs are derived, often only cover a crop growing season ( $EF_{gs}$ ), and only a few studies have noted differences ( $\Delta EF$ ) 80 between EF<sub>gs</sub> and those based on emissions measured over a whole year (EF<sub>wy</sub>): these cases 81 being, e.g. vegetable fields in Spain (Sanchez-Martin et al., 2010) and paddy rice in China (Liu 82 et al., 2016). Although the  $\Delta$ EF should not be neglected, many current regional N<sub>2</sub>O inventories 83 84 (Bouwman et al., 2002b; Cayuela et al., 2017; Stehfest and Bouwman, 2006) are based on the EFgs rather than the EFwy. None of these studies provide an assessment of whether the 85 86 difference,  $\Delta EF$ , exists ubiquitously in global agroecosystems.

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88 The difference between EF<sub>gs</sub> and EF<sub>wy</sub> depends on residual fertilizer N, and other management and environmental factors affecting nitrification and denitrification processes during fallow 89 periods. Vegetables generally have lower nitrogen use efficiency (NUE, yield N/fertilizer N) 90 91 compared to other crops (e.g., maize, wheat and paddy rice) (Garnett et al., 2013), which may contribute to high residual fertilizer-induced N<sub>2</sub>O emissions during the fallow periods, 92 accounting for 15%-50% of the annual total (Pfab et al., 2011; Sanchez-Martin et al., 2010). 93 Field drainage after rice harvest may change soil conditions from anaerobic to aerobic (Hou et 94 95 al., 2012; Peng et al., 2011; Zou et al., 2005), stimulating N<sub>2</sub>O emissions during fallow periods 96 through the inhibition of N<sub>2</sub>O reduction in denitrification (Butterbach-Bahl et al., 2013). Other management and environmental factors, such as fertilizer type, rainfall, temperature, soil 97 organic carbon content, total N content and texture, can also affect nitrification and 98 99 denitrification processes (Bouwman et al., 2002a; Butterbach-Bahl et al., 2013). However, it is not yet known whether these factors play crucial roles in any observed differences between
 EF<sub>gs</sub> and EF<sub>wy</sub>.

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The objectives of this study are to evaluate the differences between EFgs and EFwy, and to 103 identify the key factors responsible for any differences at global scale. Here we conducted a 104 global meta-analysis of 123  $\Delta$ EFs from 21 studies where each experiment contained both EF<sub>gs</sub> 105 and  $EF_{wv}$ . The overall mean  $\Delta EF$  and mean  $\Delta EF$ s for subgroups, with respect to management 106 and environmental factors, were statistically compared to zero to detect differences between 107  $EF_{gs}$  and  $EF_{wy}$ . Key factors influencing  $\Delta EFs$  were identified by identifying significant 108 109 differences of  $\Delta EFs$  within the subgroup. Finally, the implications of  $\Delta EFs$  on the estimation 110 of annual N<sub>2</sub>O emissions were quantified with respect to  $EF_{gs}$  ( $\Delta EF/EF_{gs}$ ) for upland crops and paddy rice. 111

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#### 113 2 Materials and methods

#### 114 **2.1 Data selection and collection**

To locate all relevant papers that have reported measurements of  $N_2O$  for crop growing seasons and whole years, we performed a comprehensive search on Web of Science, Google Scholar and the China National Knowledge Infrastructure database using the keywords: nitrous oxide or  $N_2O$ , non-growing or fallow, upland crops or paddy rice, and soil or fertilizer. To ensure comprehensive coverage, we also checked all references cited in the papers found.

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We selected studies from 21 peer-reviewed papers and dissertations where  $N_2O$  emissions with at least two different N application rates, including a zero control, were measured both for whole year, and for the growing season and fallow period. Studies with the following measurements were excluded: (i) measurements made in laboratories or greenhouses, (ii) measurements conducted in organic (peaty) soils where  $N_2O$ -EFs are much higher than those in mineral soils (IPCC, 2006), and (iii) measurements with the use of controlled-release fertilizers, or nitrification or urease inhibitors, which may reduce  $N_2O$  emission rates. The final dataset contains 123 paired EF<sub>gs</sub> and EF<sub>wy</sub> at 20 sites globally (Fig. 1 and Table S1).

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130 For each site, the N<sub>2</sub>O emissions and related variables were sorted into four categories: (i) N<sub>2</sub>O 131 emissions, (ii) climatic factors, (iii) soil properties, and (iv) management parameters. The N<sub>2</sub>O 132 emissions for the whole year and growing season were obtained from the studies identified. 133 The averages of these emissions by replicated measurements were then used to calculate N<sub>2</sub>O EF<sub>gs</sub> and EF<sub>wy</sub>. For the climatic factors, climatic zones based on thermal and moisture regimes 134 (cool, warm, dry and moist zones) were identified according to the locations of sites to 135 represent the variations of soil water content and temperature, following the method in Smith 136 et al. (2008) and Albanito et al. (2016). Mean annual air temperature (MAT) and mean annual 137 138 precipitation (MAP) for field sites were obtained from the original papers. Mean annual evapotranspiration (MAET) values for 1980-2010 were extracted from the Climatic Research 139 Unit (CRU) TS v. 3.23 database (https://crudata.uea.ac.uk/cru/data/hrg/). Presence of freeze-140 141 thaw cycles was characterized by the minimum soil temperature during fallow periods. When the minimum soil temperature was less than zero, we assumed that the soil water was subject 142 to freeze-thaw in winter during the fallow periods. Soil organic carbon content (SOC), pH, total 143 N content, bulk density (BD), and clay content were used to account for substrate availability 144 and soil aeration conditions, which together with climate conditions, determine rates of 145 146 nitrification and denitrification (Bouwman et al., 2013; Butterbach-Bahl et al., 2013). For the management parameters, crop type, fertilizer type, N fertilizer application rate, residue return, 147 irrigation, and fallow duration were selected, because of their known impacts on soil C and N 148 cycling and transport in the root zone. Multiple cropping seasons of different upland cereals 149

(e.g., maize-wheat rotation) in a full year were included in the category "other crops". Single
or double cropping seasons of paddy rice were both categorized as paddy rice, and the category
"vegetables" contained multiple vegetable cultivations. Information on fertilization methods
(e.g. broadcast, injection, or deep placement) and tillage practices were not available in the
original papers, so these factors were not considered for further analysis.

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Missing values of MAT and MAP (32% and 19% of total 123 AEFs, respectively) for 1980-156 2010 were extracted from the CRU climate database; BD, clay content, SOC and pH (58%, 157 36%, 7% and 0% of the total  $\Delta EFs$ , respectively) were supplemented from the 1-km 158 Harmonized World Soil Database (HWSD v1.2) (http://www.iiasa.ac.at/) using site latitudes 159 160 and longitudes. Data from CRU and HWSD were validated through observations from literature at known latitudes and longitudes (Fig. S1 and S2). Although total N could not be 161 added from external datasets, we found that missing data (7% of observations) did not impact 162 our results greatly. Details of these variables can be found in Table S2. 163

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#### 165 **2.2** ∆**EF**

166 The  $\Delta EF(\%)$  was calculated as the difference in N<sub>2</sub>O-EFs between the whole year and growing 167 season for a non-zero N application level under the same environmental and management 168 conditions. We did not average the  $\Delta EF$  for a specific site if management practices, such as 169 crops, fertilizer types, and tillage, or other critical factors were different. The N<sub>2</sub>O  $\Delta EF$  for each 170 pair of whole year and growing season was evaluated using the following equations:

$$171 \qquad \Delta EF = EF_{wy} - EF_{gs} \tag{1a}$$

172 where

173 
$$EF_{wy} = (E_{wy} - E_{0wy}) / N$$
 (1b)

174 
$$EF_{gs} = (E_{gs} - E_{0gs}) / N$$
 (1c)

and the indices *wy* and *gs* represent whole year and growing season; E is the N<sub>2</sub>O emission from the fertilized treatment (kg N ha<sup>-1</sup>);  $E_0$  is the N<sub>2</sub>O emission under zero-N control (without N application, kg N ha<sup>-1</sup>); *EF* is emission factor, %; *N* is fertilizer N application rate (kg N ha<sup>-1</sup>).

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#### 180 **2.3 Statistical analyses**

Analyses of N<sub>2</sub>O  $\Delta$ EF were conducted using statistics based on resampling. We used a 181 Kolmogorov-Smirnov test to determine if the distribution of the dataset differed from normality, 182 and given that it was not normally distributed (p<0.001), we applied the bootstrapping 183 resampling method to estimate the means of the  $\Delta EFs$ . Bootstrapping resampling (i.e., random 184 sampling with replacement of the equal size of the initial dataset repeated n=100,000 times) 185 was performed using the MATLAB bootstrapping function to generate the normal distributions 186 of the means of  $\Delta EFs$ , and then to compare 95% confidence intervals (CIs) of the means with 187 zero to identify the difference in EFs observed during whole year and growing season. 188 Differences between subgroups of potential factors were then tested using bootstrap confidence 189 intervals and analysis of variance (ANOVA) on bootstrapped values for two and multiple 190 191 subgroups, respectively.

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We removed 1 outlier  $\Delta EF$  with the largest value (1.03%) from further analysis due to its undue influence on the means of subgroups, since it was nearly 7 times larger than the standard deviation of the dataset. The remaining 122  $\Delta EFs$  were then categorized into 16 groups: crop type, climatic zone, MAT, freeze-thaw cycles, MAP, potential net water input (MAP–MAET), SOC, pH, total N content, BD, clay content, fertilizer type, N application rate, residue return, irrigation, and fallow duration. For the groups with continuous variables (e.g. total N content and SOC),  $\Delta$ EFs were split into two subgroups based on the responses of  $\Delta$ EFs to the factors.

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We used a contingency table to show the interrelation between different environmental factors to avoid assigning the same influence to two or more factors. For each pair of soil properties or climatic factors, we calculated the phi coefficient ( $\phi$ ), the degree of association between two variables of two categories. In the contingency table,  $\phi = \pm \sqrt{\chi^2/N}$ , where  $\chi^2$  is from Pearson's  $\chi^2$  test, and the total number of observations, N, was given.

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#### 207 2.4 Impact of ΔEF on N<sub>2</sub>O-EF

The percentage of  $\Delta EF$  of the mean  $EF_{gs}$  was used to indicate the relative impact of  $\Delta EF$  on EF<sub>gs</sub> for N<sub>2</sub>O emission inventories. We estimated mean  $\Delta EFs$  and their 95% CIs by the bootstrapping resampling method for paddy rice and non-vegetable upland crops. The mean EF<sub>gs</sub> was then calculated based on the data collected in this study. Moreover, we used the proportional upper and lower boundaries of the 95% CI to estimate the uncertainty of the mean EF<sub>gs</sub>. To compare EF<sub>gs</sub> with other studies, N<sub>2</sub>O datasets from Cayuela et al. (2017) and Akiyama et al. (2005) were also collected.

#### 216 **3 Results**

An analysis of all data together showed significantly greater EF values for a whole year than for a growing season.  $\Delta$ EFs were positive in 86% of the data and negative in only 14% (17 cases) under certain circumstances (Text S1). Based on the bootstrap resampling, both the mean (0.08) and median (0.03) of the overall  $\Delta$ EFs were significantly positive (p< 0.001), with 95% confidence intervals of 0.06-0.11 and 0.02-0.05, respectively. Removing 1 outlier from the dataset decreased the mean from 0.081 to 0.073 but had no significant effect on the median value (Fig. 2).

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#### 225 **3.1 Impact of crop type on ΔEF**

Three main cereal crops (paddy rice, n=35; maize, n=23, and wheat, n=33) dominated other 226 crops (vegetables, n=10; legume, n=4, and other upland crops, e.g. barley, maize and wheat 227 rotations, n=21) on the perspective of  $\Delta EF$  data availability. Except for legume crops, for which 228 the data were insufficient, all crops showed significant positive  $\Delta EF$  values (p<0.01). However, 229 230 differences among crop types showed that mean  $\Delta EF$  for paddy rice was significantly larger (p<0.05) than that for all upland crops, except for vegetables (Fig. 2). The mean  $\Delta EFs$  for paddy 231 rice (0.11) and vegetables (0.19) were about 2 to 7 times those for maize (0.03), wheat (0.07)232 233 and other upland crops (0.04).

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#### **3.2 Impact of climate on** $\Delta EF$

The  $\Delta EFs$  for the moist climatic zones (warm-moist and cool-moist) were significantly (p<0.001) positive (Fig. 3a). The cool-moist zone had the highest mean  $\Delta EF$  (0.10), followed by warm-moist (0.08) and warm-dry (0.01). Significant (p<0.05) differences were also found between moist (cool-moist and warm-moist) and dry (warm-dry) zones. The averaged  $\Delta EF$  for moist regions was 0.09, ~6 times more than that of the dry region. The subgroup for water condition also had significantly (p<0.001) positive  $\Delta EF$  values. The  $\Delta EF$  was much larger when the MAP reached 600 mm per year, however the effect of the difference between precipitation and evapotranspiration (MAP-MAET) was not significant. Higher ( $\geq 15^{\circ}C$ ) or lower (<15°C) MAT, and the occurrence of freeze-thaw cycles during the fallow period, showed no significant impacts on the magnitude of the  $\Delta EF$  values (p<0.001).

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#### 247 **3.3 Impact of soil properties on** $\Delta EF$

Among soil conditions,  $\Delta EFs$  were all significantly higher (p<0.001) than zero in relation to SOC, pH, total N content, BD and clay content. The  $\Delta EFs$  were also higher where SOC contents were relatively high (SOC  $\geq$ 1.3%), and where soil was acidic (pH <7; Fig. 3b). The total N content, BD, and clay content had no significant impact on the  $\Delta EF$  at the 95% confidence level.

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#### 254 **3.4 Impact of management on ΔEF**

All N fertilizer types (Urea (U), Organic (O), mixture of synthetic and organic fertilizers (OS) and other synthetic fertilizers (other SNs)) and application rates had significantly positive impacts on  $\Delta EFs$  (p<0.05, Fig. 3c). The mean  $\Delta EFs$  for U (0.08) and other SNs (0.10) were ~3 times more than that for O (0.03) and OS (0.04), showing significant (p<0.05) differences. Although the mean  $\Delta EF$  increased with increasing N fertilizer application rate, the differences between them were not significant (p>0.05).

For other agricultural management, i.e., residue management, irrigation and length of fallow 262 period, the mean  $\Delta EFs$  for all subsets were significantly positive (p<0.05) (Fig. 3d). The mean 263  $\Delta EF$  was higher (p<0.05) for residue return (0.12) after harvest than that for residue removal 264 (0.03). Irrigated cropland had a higher mean  $\Delta EF$  (0.09, n=75) compared with the mean of all 265 data, whereas rain-fed cropland (n=7) had substantially smaller mean  $\Delta EF$  (0.01). Among three 266 categories of fallow duration, the mean  $\Delta EF$  grew significantly (p<0.05) with increasing length 267 268 of fallow periods and significantly (p<0.05) positive correlation was found between the  $\Delta EF$ and the duration of fallow period. The mean  $\Delta EF$  for fallow periods of  $\geq 200$  days (0.11) was 269 270 ~2 and ~6 times higher than the mean  $\Delta EF$  for fallow periods of 100-200 days and <100 days.

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Relatively greater factor associations ( $\phi$ >0.4 or <-0.4) were primarily found between climatic zone and other environmental factors in a contingency table (Table S3). Other large associations were between total N and SOC (0.64), between pH and MAT (-0.65), pH and MAP (-0.44), and between total N and freeze-thaw cycles (0.58). High levels of relatedness were also found between other climatic factors, e.g., MAT and MAP (0.48), MAT and freezethaw cycles (-0.57), and MAP and MAP-MAET (0.86).

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#### 279 **3.5 Impact of \Delta EF on N<sub>2</sub>O-EF**

Paddy rice and non-vegetable upland crops both had positive (p<0.05) mean proportional  $\Delta EFs$ and corresponding 95% CIs (Fig. 4). The mean proportional  $\Delta EF$  for paddy rice was 30% (95% CI: 16%-45%, n=31), nearly 3 times that for non-vegetable upland crops (11%, 95% CI: 7%-14%, n=81). The proportional upper and lower boundaries of 95% CI of mean EF<sub>gs</sub> in this study were 26% and -23% for upland crops (n=81), and 49% and -40% for paddy rice (n=31),
respectively.

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#### 287 4 Discussion

Our results show that the N<sub>2</sub>O-EF for a whole year is greater than that for the growing season; that is, positive mean  $\Delta$ EFs were found for the overall dataset and for most subgroups by crop type, climatic factor, soil property, fertilization practice, and other management practices (Figs. 2 and 3).

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#### 293 **4.1 Effect of crop type on** $\Delta EF$

Vegetables showed significantly greater differences between whole year EF and growing 294 season EF compared to other upland crops (e.g., wheat, maize, legumes) (Fig. 2). The nitrogen 295 use efficiency (NUE; yield N/fertilizer N) for vegetables has been reported to be 14%, 296 297 substantially lower than those for wheat (42%), maize (46%), paddy rice (39%), legume (80%) 298 and other cereal crops (53%) (Zhang et al., 2015). This is related to high fertilizer N inputs for intensive cropping vegetable systems (Li et al., 2017; Zhang et al., 2012). Globally, vegetables 299 use 7% of global synthetic N fertilizer (Patrick et al., 2017), but account for only ~4% of 300 301 harvested cropland area (FAO, 2019), leading to an application rate that is more than 30% than for other crops (Fig. S3). As reported by Gerber et al. (2016), vegetables had a slightly higher 302 303 EF than other crops (e.g., 2% and 12% higher than maize and wheat, respectively). The high N application rate and the low NUE would be expected to lead to more N substrate being available 304 for N<sub>2</sub>O production during the fallow period (see Eq. S1 and Text S2), which is supported by 305 a significant and positive relationship (r=0.3) between N application and N<sub>2</sub>O emissions during 306 the fallow season found in this study. 307

Paddy rice showed a significantly larger  $\Delta EF$  compared with other non-vegetable upland crops 309 (Fig. 2). Given that N application rates for non-legume crops are similar (210±28 kg N ha<sup>-1</sup>, 310 mean  $\pm$  standard deviation) in our dataset, the magnitude of the  $\Delta EF$  is determined only by 311 312 residue fertilizer-induced N<sub>2</sub>O emissions, i.e., the difference in N<sub>2</sub>O emissions between fertilized and unfertilized plots during the fallow period (Eq. S1 and Text S2). The residual 313 fertilizer-induced N<sub>2</sub>O flux during the fallow period was 6  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup> (n=32) for paddy rice, 314 which is about twice that of wheat (n=33) and about 5 times that of maize (n=23) (Table S4). 315 The N<sub>2</sub>O fluxes for these upland crops decreased significantly from the growing season (23 µg 316 N m<sup>-2</sup> h<sup>-1</sup>. n=82) to the fallow period (9  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, n=82), which may be due to lower N 317 availability in soils, while the N<sub>2</sub>O fluxes for paddy rice did not decrease during fallow period 318 (19 vs. 16  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, p=0.52). This coincides with a change in soil water and oxygen 319 320 conditions after flood drainage that favor N<sub>2</sub>O emissions during the non-growing period (Majumdar, 2013; Wang et al., 2013; Zheng et al., 2010). N<sub>2</sub>O emissions in paddy rice systems 321 are impacted greatly by flooding and drainage cycles. For example, the meta-analysis of 322 Akiyama et al. (2005) reported that the practice of midseason drainage led to a larger EF than 323 continuous flooding. Though paddy rice is cultivated mostly in regions with a moist climate 324 325 and upland crops are grown both in moist and dry areas, our results do not suggest that the higher  $\Delta EF$  for paddy rice resulted from climatic factors. Instead, we propose that it was most 326 likely due to residual N fertilizer, since the residual fertilizer-induced N<sub>2</sub>O flux for paddy rice 327  $(6 \mu g N m^{-2} h^{-1}, n=32)$  was still significantly (2 and 4 times) greater than those for wheat (n=32) 328 and maize (n=9) in moist areas (Table S4). For other factors with high associations ( $\phi$ >0.4 or 329 <-0.4, Table S3), such as pH, MAT and freeze-thaw cycles, significantly larger residual 330 fertilizer-induced N<sub>2</sub>O flux was also found for paddy rice than for other non-vegetable crops 331

332 (Table S4). Hence water management factors, such as flooding and drainage, are dominant over 333 other factors in affecting the  $\Delta EF$  for paddy rice.

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#### 335 **4.2 Effect of climate on** $\Delta EF$

336 Our results showed that precipitation was the dominant climatic factor over temperature, showing significant differences in  $\Delta$ EF: moist areas had greater  $\Delta$ EF than dry areas (Fig. 3a). 337 Higher precipitation was found to significantly promote greater N<sub>2</sub>O emissions in the fallow 338 season and consequently resulted in greater  $\Delta EF$  values in this study, as evidenced by the 339 significant and positive relationship (r=0.3) between the amount of rainfall during the fallow 340 341 period and the  $\Delta EF$  (Fig. S4). Higher precipitation is associated with higher soil water content, which is known to be a major driver of N<sub>2</sub>O emissions, by regulating oxygen availability to 342 343 microbes in the soil (Butterbach-Bahl et al., 2013; Davidson et al., 2000; Song et al., 2019). Rising soil water content has been reported to increase N<sub>2</sub>O emissions (Smith et al., 2003; 344 Bateman and Baggs, 2005), before extreme soil anaerobic conditions favor the reduction of 345 346 N<sub>2</sub>O through denitrification (Butterbach-Bahl et al., 2013; Davidson et al., 2000). A recent meta-analysis conducted by Xia et al. (2018) also reported a positive relationship between 347 precipitation and N<sub>2</sub>O emissions. Although MAT and MAP-MEAT had relatively large 348 associations with MAP ( $\phi$ >0.4 or <-0.4, Table S3), only MAP had a significant impact on  $\Delta$ EF. 349

- 350
- 351 **4.3 Effect of soil properties on**  $\Delta EF$

Our results show that SOC and pH have important effects on  $\Delta$ EFs (Fig. 3b), which appear to be related to their impacts on N<sub>2</sub>O-EF from residual fertilizer N during non-growing periods. A positive relationship between N<sub>2</sub>O emissions and SOC content was also found in other global

meta-analyses (Bouwman et al., 2002a; Charles et al., 2017). Lower soil pH is generally known 355 to depress the activity of N<sub>2</sub>O reductase enzymes and the reduction of N<sub>2</sub>O to N<sub>2</sub> in the 356 denitrification process (Bakken et al., 2012; Čuhel et al., 2010; Wang et al., 2018). Soil pH 357 may also affect other biotic or abiotic processes (e.g., nitrification and chemical denitrification) 358 and the microbial community and may thereby affect N<sub>2</sub>O emissions, but the mechanisms 359 remain unclear (Wang et al., 2018). A negative relationship between pH and soil N<sub>2</sub>O emission 360 361 has been observed in laboratory studies with soils from acidic tea fields (Tokuda and Hayatsu, 2001), paddy rice fields (Shaaban et al., 2014; Shaaban et al., 2018) and in a recent meta-362 analysis (Wang et al., 2018). The association between SOC and pH was relatively low ( $\phi = -0.1$ , 363 Table S3) in this study, suggesting that SOC and pH may be independently affecting N<sub>2</sub>O 364 emission during fallow periods. 365

366

#### 367 **4.4 Effect of management on** $\Delta EF$

Our results showed that the  $\Delta EF$  under organic fertilizer (livestock manure) application was 368 significantly lower than that under urea application (Fig. 3c). As discussed in Section 4.1 and 369 Text S2, it appears that the residue fertilizer-induced N<sub>2</sub>O flux during fallow period has the 370 371 greatest impact on the  $\Delta EF$ , given that N application rates for organic fertilizer and urea in our dataset were comparable (166 v.s. 172 kg N ha<sup>-1</sup>, p=0.16). The residue fertilizer-induced N<sub>2</sub>O 372 flux for organic fertilizer (2  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, n=32) was one third that for urea application (n=45). 373 Similarly, a meta-analysis conducted by Xia et al. (2017) reported that, compared to urea 374 application, organic fertilizer also reduced N<sub>2</sub>O emissions by 11.4%, attributed to the 375 376 significant promotion of microbial N immobilization (by 36.4%), thereby reducing the availability of N for N<sub>2</sub>O productions (Zhou et al., 2016). 377

The combination of crop residue return with synthetic N fertilizer led to a significantly higher 379  $\Delta$ EF compared to a single synthetic N fertilizer application at comparable N application rates 380 (194 v.s. 186 kg N ha<sup>-1</sup>, p=0.78) (Fig. 3c). Straw degradation provides additional N substrates 381 for nitrification and denitrification, which stimulates N<sub>2</sub>O emissions in the fallow crop season 382 (Liu et al., 2014). The  $\Delta EF$  for paddy rice with crop residue return (0.15%, n=20) was three 383 times greater than that for non-vegetable upland crops with crop residue return (n=31). The N<sub>2</sub>O 384 fluxes for upland crops decreased significantly from the growing season (29  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, 385 n=31) to the fallow period (16  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, n=31), which may be due to lower availability of 386 N in soils, while the N<sub>2</sub>O fluxes for paddy rice did not decrease during the fallow period (14 387 v.s. 10  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, p=0.33). It is coincident with the change of soil water and oxygen 388 conditions that favor N<sub>2</sub>O emissions resulting from drainage after harvest in paddy fields 389 390 (Akiyama et al., 2005; Majumdar, 2013; Wang et al., 2013; Zheng et al., 2010). The metaanalysis of Xia et al. (2018) reported that the return of crop residue significantly decreased N<sub>2</sub>O 391 emissions (by 17.3% in the flooded rice-growing season while it increased emissions by 21.5% 392 393 for upland crops, supporting the findings reported here.

394

Fallow duration is significantly and positively correlated to  $\Delta EF$  (Fig. S5 and Fig. 3d), because a longer non-growing period means a shorter growing season within a year, which also represents more residual fertilizer N available for N<sub>2</sub>O production from both nitrification and denitrification during fallow periods.

399

400 4.5 Effect of  $\Delta EF$  on N<sub>2</sub>O-EF

The 95% CIs of mean  $EF_{gs}$  in this study were similar to those from two meta-analyses for 401 growing-season Mediterranean upland crops (n: 186; upper: 20%, lower: 26%) (Cayuela et al., 402 403 2017) and global paddy rice (n=50; upper: 33%, lower: 31%) (Akiyama et al., 2005) (Fig. 4). The mean proportional  $\Delta EFs$  for upland crops and paddy rice were 11% and 30% (Figs. 4a and 404 b), which means that an EF<sub>gs</sub>-based emission inventory, using growing season measurements 405 only, could underestimate N<sub>2</sub>O emissions by one tenth and one third, respectively. Moreover, 406 407 for paddy rice, the mean proportional  $\Delta EF$  and its upper boundary of 95% CI (45%) were close to, or even exceeded, the upper boundaries of the mean EFgs from this study (49%) and 408 409 Akiyama et al. (2005) (33%). Neglecting N<sub>2</sub>O emissions from the non-growing season will 410 lead to an underestimation of cropland  $N_2O$  emissions for both crop types, especially for paddy rice, so emission mitigation for non-growing periods also needs to be considered. 411

412

#### 413 **4.6 Limitations**

The main limitation of the analysis is the lack of available whole-year measurements, because 414 most measurement campaigns have focused only on cropping seasons. Geographical coverage 415 is also an issue; with more  $\Delta EFs$  from Africa, South America and East Europe, we would be 416 417 better able to capture the magnitude and important factors for  $\Delta EF$  with higher confidence. Additional studies are especially needed in cool-dry and warm-dry climatic zones, for 418 vegetables and legumes and under different irrigation regimes. With more  $\Delta EFs$  for vegetables, 419 420 interdependences of environmental and management factors can be tested to determine their relative importance. In addition, more studies with two or more non-zero N application levels 421 422 are required for studying the impacts of N input rate on  $\Delta EF$ . Soil amendments, such as controlled-release fertilizer and nitrification inhibitors, may potentially lead to an increase in 423 fallow N<sub>2</sub>O emissions, due to the prolonged release of nitrogen. Further knowledge of the 424

factors controlling the differences in EF between whole years and growing seasons, and their
magnitude, is crucial for reducing the uncertainties of N<sub>2</sub>O inventories and the corresponding
greenhouse gas balance of croplands.

428

#### 429 **5** Conclusions

This meta-analysis showed that the inclusion of non-growing season N<sub>2</sub>O emission 430 significantly increased cropland N<sub>2</sub>O-EF, indicating that residual fertilizer-induced N<sub>2</sub>O 431 432 emission during the non-growing season cannot be neglected for national inventories. In particular, ignoring emissions from the non-growing season can underestimate the N<sub>2</sub>O-EF by 433 30% for paddy rice and by ~10% for non-vegetable upland crops. Areas with high precipitation, 434 high soil organic carbon content, or low pH experience higher risks of residual fertilizer-435 induced N<sub>2</sub>O emissions. For national cropland N<sub>2</sub>O emission estimates and mitigation 436 strategies, frequent measurements of N<sub>2</sub>O emission should be taken both during the crop 437 growing and non-growing periods. In the future, attention should be paid to the fate of residual 438 439 fertilizers and their effects on the environment.

### **Declaration of interests**

442 The authors declare no competing interests.

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### 452 Author contributions

- 453 Z.S., P.S. and M.A. conceived and designed the research; Z.S. and M.A. collected data; Z.S.
- 454 performed the analysis; Z.S., M.A., M.K., F.A., and L.X interpreted the results; Z.S. wrote the
- 455 paper with contributions from M.A., M.K., L.X and P.S.; All revised the paper.

### 456 Supplementary material

457 Additional Supplementary Information can be found in the online version of this article.

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606 Figure captions

Fig.1 Map showing the locations of experimental sites and numbers of observations used
 in this paper (123 paired N<sub>2</sub>O EF<sub>gs</sub> and EF<sub>wy</sub> for whole year and growing season at 20
 sites). Green area represents global croplands.

610

611Fig.2 Overall ΔEF and ΔEF grouped by crop types. Data are presented as mean ± SEM,612with n noted at the base of each bar. Asterisks indicate significant differences from zero613(\*\*\*p< 0.001; \*\*p < 0.01; \*p<0.05). Different letters indicate significant differences between</td>614mean ΔEFs for groups within each category. "Others" represent upland crops except wheat,615maize and legume.

616

Fig.3 AEFs by climatic factors (a), soil attributes (b), fertilization (c), and other 617 managements (d). Data are presented as mean  $\pm$  SEM, with n noted at the base of each bar. 618 Asterisks indicate significant differences from zero (\*\*\*p< 0.001; \*\*p < 0.01; \*p<0.05). 619 Different letters indicate significant differences between mean  $\Delta EFs$  for groups within each 620 category. <sup>1</sup>: The minimum soil temperature is presumably an indicator for the occurrence of 621 freeze-thaw cycles: if the minimum soil temperature is below 0 °C, then the freeze-thaw cycle 622 is assumed to occur.<sup>2</sup>: The potential net water input is defined as the difference between mean 623 annual precipitation and evapotranspiration. 624

626 Fig.4 Impact of ΔEF on EF<sub>gs</sub> for non-vegetable upland crops (a) and paddy rice (b). The 627 effect of ΔEF is represented by the ratio of its mean and 95% confidence interval (CI)

- boundaries to the mean  $EF_{gs}$  in this study (excluding 1 outlier). 95% CIs of the mean  $EF_{gs}$  in
- this study were in comparison with those from Cayuela et al. (2017) and Akiyama et al. (2005).



632 Fig.1



634 Fig.2



636 Fig.3





638 Fig.4