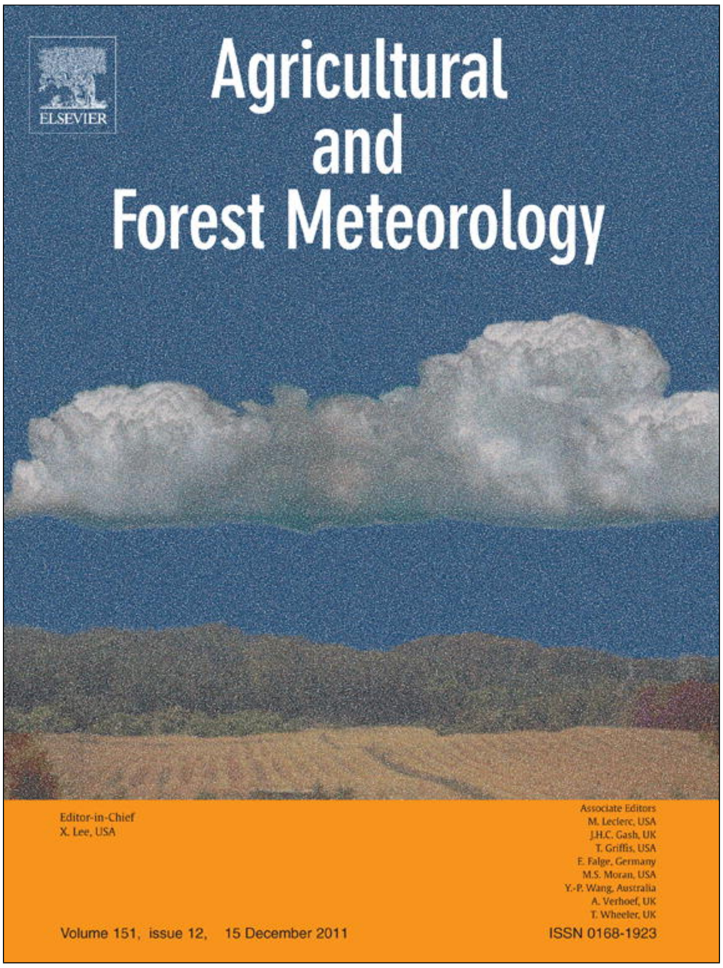

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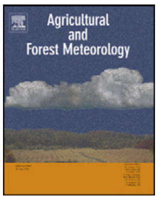

Gap-filling techniques for the annual sums of nitrous oxide fluxes

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ABSTRACT

The full accounting of greenhouse gas emissions is only possible when the three key contributors (CO_2, CH_4 and N_2O) are continuously measured throughout the observation period. While some field techniques such as eddy covariance or automatic closed chamber measurements have attempted near-continuous observations, it is always beyond the control of the experimentalist to ensure perfect continuity. Therefore, the final time series of fluxes will inevitably have periods without reliable values (i.e., gaps) and will need to be gap-filled. While there is abundant literature on methodologies for gap-filling CO_2 fluxes, there is no literature on the gap-filling methods for trace gases such as nitrous oxide. We investigate three general approaches for gap-filling nitrous oxide time series: linear extrapolation, moving average and look-up tables. A five-year time-series of eddy-covariance measurements from an intensive grassland site in South-Western Ireland was used as an example. The amount of gaps varied significantly from year to year. The single-year look-up table technique produced consistently good results even when long gaps were present in the time series. In some years, the simpler annual extrapolation technique performed equally well. It is essential that this work be extended with more complex methods, and that methods in this paper are further evaluated under different environmental conditions. We believe that these methods and analysis could also be applied to other trace gas gap-filling (e.g., CH_4) which have similar intermittent patterns of emission.

1. Introduction

Nitrous oxide (N_2O) is a powerful greenhouse gas (GHG) with a global warming potential of 298 relative to CO_2 over a 100-year time horizon (Forster et al., 2007). Additionally, N_2O is involved in the reduction of the stratospheric concentrations of ozone (Crutzen, 1970). Traditionally, monthly and annual sums of N_2O emissions are estimated from intermittent observations. Gap-filling, therefore, would have to be used to produce a robust and reliable balance of N_2O emissions. There is, however, little or no information in the literature regarding any gap-filling approaches, and to the best of our knowledge no comparison study similar to those performed for CO_2 (Falge et al., 2001; Moffat et al., 2007) has been published.

Leahy et al. (2004) used the modified moving daily average method to calculate the annual sum of N_2O emissions. The instantaneous fluxes (30-min averages) were aggregated into daily values by linear extrapolation or in case of less than 12 values available (out of possible 48) were gap-filled with the moving average algorithm using a 5-day window. Such an approach, while appearing reasonable, was not justified by any assumption regarding typical patterns of emission at the site, or as reported in literature. Little explanation was provided in the earlier paper by Scanlon and Kiely (2003) that used a similar approach, with linear interpolation instead of a moving average.

A number of approaches of various complexity are available for CO_2 gap-filling (Falge et al., 2001; Moffat et al., 2007; Reichstein et al., 2005). Carbon dioxide fluxes are relatively straightforward to gap fill as they not only tend to follow a diurnal cycle but also have seasonal and inter-annual similarities. Furthermore, there is a much deeper understanding of the drivers of CO_2 fluxes and both empirical and mechanistic models provide stronger support to the CO_2 gap-filling methods. Due to natural differences between carbon dioxide and nitrous oxide, it would not be possible to directly apply all of the CO_2 gap-filling methods to the fluxes of N_2O. Key features of N_2O fluxes are their intermittent occurrence and the presence of burst events; and, therefore, some techniques such as mean diurnal variation would be severely limited by a particular flux-measurement methodology and could be restricted to only certain seasons. Some of the approaches, however, can be transferred onto the N_2O data set with minor modifications. For example, the look-up table technique (Falge et al., 2001; Reichstein et al., 2005) can be applied for N_2O gap-filling with appropriate parameters.

The existing methods can be classified as sequential—based on the location of the gap within the time series (e.g., mean diurnal); based on environmental-variables (look-up tables); governed

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by the environmental processes (non-linear regressions, biosphere models); black box (artificial neural networks). Of these groups, only the environmental-variables-based approach can be translated in a straightforward way for N_2O gap-filling.

The aim of this paper was to investigate possible gap-filling approaches for the purpose of calculating the annual emission of N_2O. We limited this study to approaches that are straightforward to implement and required only the most-widely employed environmental characteristics. We also hope that this work will prompt further discussion and development of gap-filling techniques for trace gases other than CO_2, particularly now that there are a number of N_2O/CH_4 data sets becoming available within the GHG flux community.

2. Methods

2.1. Site description and flux measurements

The study site is located near the village of Donoughmore, Co. Cork, in South-Western Ireland. The geographical coordinates are 51°59′N, 8°45′W, and the mean elevation is 187 m (above sea level). Most of the site is poorly drained with a small area prone to winter water logging. The soil is gleyic cambisol with an annual water table ranging from approximately 0.6 to 2.5 m below the surface. The topsoil is rich in organic matter to a depth of about 15 cm (about 10% organic matter), overlying a dark brown B horizon of sand texture (Scanlon and Kiely, 2003). Within the 0–10 cm layer the soil bulk density was 1.02 g cm^{-3}, pH was 6.7, total soil organic carbon (SOC) was 4.5%, and total soil nitrogen was 0.35% (C:N ratio of 13). Averaged over the top 10 cm, the soil porosity was 0.60, the saturation moisture level was 0.57, the field capacity was 0.32, and the wilting point was 0.12 m^3 m^{-3}. The typical land use in the region is grassland with a mix of dairy and beef cattle grazing and grass harvesting for silage and hay. The dominant plant species at the site is perennial ryegrass (*Lollium perrene* L.) with a minor presence of clover. Details of fertilizer application over the study period are presented in Kim et al. (2010).

The eddy-covariance setup used to measure the N_2O flux consisted of a CSAT3 3-D sonic anemometer (Campbell Sci., USA) and a tuneable diode laser trace-gas analyser (TGA 100A, Campbell Sci., USA). Both the anemometer and the N_2O intake were mounted at 6 m height, and the data were collected with a CR1000 data logger (Campbell Sci., USA) at a frequency of 10 Hz. The ground instrumentation included soil temperature probes at 5 cm, time-domain reflectometry (TDR) probes for soil moisture at the same depth and generic tipping-bucket rain gauges with a resolution of 0.1 mm.

The 10 Hz raw data were processed into 30-min average fluxes according to the algorithm described by Mishurov and Kiely (2010). The time series of daily fluxes and the corresponding rainfall and soil temperature were reported by Mishurov and Kiely (2010, Fig. 2). Only the grassland sector time series was used in this study and its duration was five years, 2004–2008.

2.2. Gap-filling techniques

The amount of gaps varied significantly over the observation period (Fig. 1). Generally, the values were well within the range of values reported for other eddy-covariance systems (Falge et al., 2001). The exceptionally high percentage of gaps in 2006 (85.1%) was due to the intermittent data logger failures that amounted to 10 months (including 3 months in 2007). Some of the gap-filling techniques were not capable of dealing with such extremely large gaps; it was, therefore, expected that as the gaps occurred in the warmer and more agriculturally intensive periods, the final annual sums for 2006 would underestimate true emission values and appear smaller than the annual sums obtained with other techniques.

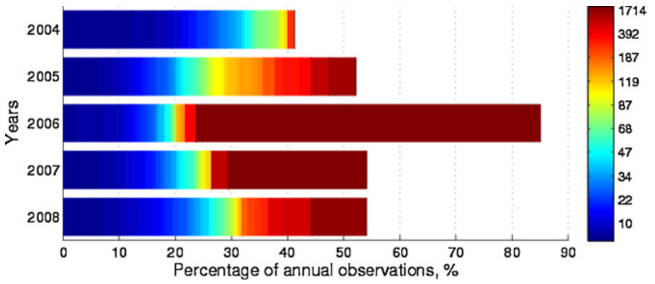

Fig. 1. Gap distribution by gap length. Colours represent the length of individual gaps (in half-hourly values); the width of segments corresponds to the cumulative percentage of gaps of a certain length.

Three fundamentally different gap-filling approaches were investigated: (1) linear extrapolation; (2) moving average; and (3) look-up tables.

2.2.1. Linear extrapolation

Linear extrapolation is a method of sequential gap-filling that is routinely used in the calculation of the annual fluxes based on infrequent chamber measurements and was reported for some eddy covariance fluxes (Allard et al., 2007, infrequent chamber measurements; Di Marco et al., 2004, eddy covariance; Wu et al., 2010, automated chambers). Because of the higher temporal resolution of EC measurements (i.e., 30 min), extrapolation of the existing flux values leads to less uncertainty than manual closed-chamber measurements which are typically conducted once per week. The essence of this method can be expressed as follows:

$$F = \frac{N}{n}\sum_{i}^{n} f_i \tag{1}$$

where F is the gap-filled flux for a certain period (e.g., a day), n is total number of periods (e.g., 30-min periods) with good flux values, N is the total number of averaging periods in a gap-filled value, f_i are the values of instantaneous flux for the good periods. It can also be described as gap-filling individual values (30-min averages) with the averages of the good values for a given period. Three temporal scales were used to evaluate the performance of gap-filling: daily, monthly and annual; correspondingly, these techniques were abbreviated as DE, ME, AE. In each case, gaps were filled with the average of good values for that particular period and the gap-filled time series was summed to obtain the final annual flux. If no good values were registered in a period, no further gap-filling was performed. Thus, it is possible that some gaps would remain unfilled when using daily or monthly extrapolation techniques.

2.2.2. Moving average

The moving average technique (MA) was used by Leahy et al. (2004). Similar to the linear extrapolation (at daily and monthly scales) this technique might not gap-fill some of the longer gaps (those that are longer than an arbitrarily declared window length). Due to a similar operating time frame, it might be expected that the performance and annual flux values would be close to those of the daily extrapolation technique. This technique was included in the study as an EC-specific gap-filling method previously used in a published paper (Leahy et al., 2004).

2.2.3. Look-up table

The look-up table (LUT) approach is a method that assigns flux in the gap period according to the environmental and management conditions prevailing at the time of the gap. This approach con-

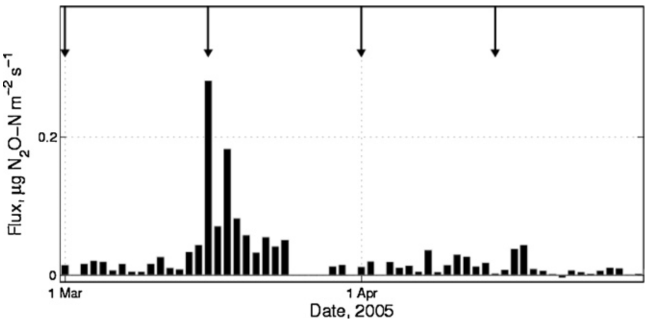

Fig. 2. Time series of fluxes of nitrous oxide (bars) and fertilizer applications (arrows) for a period March–April 2005.

sists of creating a table with the flux values binned, based on the corresponding values of the external parameters (e.g., soil temperature, rainfall, soil moisture, etc.). The determination of the relevant parameters and their critical values is a crucial step if this technique is to be successful. In this study the following parameters were used to create the table:

- cumulative rainfall in the preceding 12 h with three categories: no rainfall, 0–2 mm and 2 mm or more. Here we use cumulative rainfall as a proxy value for the water-filled pore space, the soil moisture indicator that was shown to be closely related to the N_2O emissions, but is typically harder to obtain and standardise due to high variation by depth;
- soil temperature at 5 cm depth with four categories, separated by three quartiles: below 25th percentile, above 25th percentile and up to median, above median and up to 75th percentile, and above 75th percentile;
- a binary flag for inorganic and organic (animal slurry, manure) nitrogen fertilizer applications in the preceding 5 days. Fig. 2 provides an example of a non-gap-filled daily flux time series along with the time series of fertilizer applications, indicating a close relationship in some cases.

In this way, 24 groups could be arranged from the available data set of existing environmental parameters. The choice of the particular parameter categories was based on our earlier unpublished work at the site and may vary for other sites. For each group, the corresponding flux values were averaged and the mean was used to fill the gap periods.

With the data set available to us, there were two possible ways of constructing the look-up table: single-year or multi-year LUT (LUT_sy and LUT_my, respectively). The application of the later type is limited by the assumption that no significant changes in the LUT-defining parameters occurred. Our ability to make such an assumption depends on whether the defining parameters (rainfall, temperature and fertilizers) were stationary over the study period. It is known that the amounts of N fertilizers decreased over the years (Kim et al., 2010); however, because only a binary flag for fertiliser application was used, and there is no reason to expect any trend in the two other parameters beyond normal variations, we consider these parameters stationary. This assumption could also be tested *a posteriori* based on the results of the gap-filling by both techniques: if the result of LUT_my gap-filling are significantly skewed compared to LUT_sy, one can conclude that the assumption did not hold.

Therefore, a total of six techniques were investigated in this study: daily, monthly and annual extrapolation, moving average, single- and multi-year look-up tables.

Table 1
The annual sums obtained with different gap-filling techniques.

Method	Annual sums, kg N_2O-N ha^{-1}				
	2004	2005	2006	2007	2008
Not gap-filled	3.55	2.07	0.56	2.74	1.79
Extrapolation					
Daily	6.41	3.94	1.94	4.62	2.95
Monthly	6.15	4.24	2.51	4.67	3.33
Annual	6.05	4.31	3.77	5.97	3.89
Moving average	6.23	4.51	1.12	4.48	3.04
LUT					
Annual	6.20	4.17	3.82	5.25	3.74
Multi-year	5.60	4.50	4.88	5.25	4.45

2.3. Statistical evaluation of the gap-filling techniques' performance

The statistical evaluation of the performance of each of the six techniques was conducted according to the approach described in Moffat et al. (2007). The total of 50 scenarios was separated into five groups by the duration of the individual gap periods: a single half-hour gap (very short), short (4 h), medium (32 h) and long (12 days), and the mixed scenario (400 very short, 50 short, 6 medium and 1 long gaps). The total amount of generated gaps in each scenario was about 10% of the year (Moffat et al., 2007, see Appendix A.1 for details). The artificial, generated gaps were superimposed on the existing time series without regard for natural gaps. In the case of multi-year LUT, each scenario was replicated for each year.

For each of the 50 scenarios, the following statistical parameters were considered: the coefficient of determination (R^2), the absolute and relative root mean square error (RMSE), the mean absolute error (MAE) and the bias error (BE). The long gaps cannot be filled with the techniques that operate on a daily time scale (e.g., DE or MA); hence these statistics were not calculated for long-gaps scenarios.

3. Results and discussion

3.1. Annual emission estimates

The gap-filled annual sums, presented in Table 1, could be considered estimates of the true annual flux. It is notable that with the exception of 2006, the overall agreement between the six techniques is reasonable. The significant differences in amount and distribution of gaps year over the year prevent us from using a single rule to evaluate these estimates. In 2004, the year with the lowest gap count, estimates agree well with the exception of LUT_my sum (5.6 kg N_2O-N ha^{-1}) which differed most from the median (6.2 kg N_2O-N ha^{-1}) of all 2004 values. It seems that the median value might serve as the best approximation of the true annual flux for 2004. Considering the range of good estimates and the low count of gaps, we think that the 0.1 kg N_2O-N ha^{-1} may well characterise the uncertainty of the best approximation. In the year 2005, no significant outliers were apparent and as a result both average and median agree very well; thus their value (4.3 kg N_2O-N ha^{-1}) serves as the best approximation. The gap percentage was, however, larger in 2005 than 2004, therefore the uncertainty of this approximation is likely to be larger, and we consider that 0.2 kg N_2O-N ha^{-1} (about half the size of range of values) might well describe the uncertainty estimate. In 2006, the year with the largest and most continuous gap, the variation between the techniques is large. However, the values obtained with DE, ME and MA techniques appear to be unreasonably low; when those value were excluded, the average of three remaining estimates (4.2 kg N_2O-N ha^{-1}) could serve as the best approximation. With only a small fraction of the time series available for gap-filling as good values, the uncertainty of the above approximation would need to comfortably accommodate the range of estimates and we, therefore, put it at 0.7 kg N_2O-N ha^{-1}. In 2007, the first three months of year had a prolonged gap and this period had little or no agricultural activity with the observed air temperature below 8 °C. Therefore, extrapolating values from other periods of the same year as annual extrapolation technique might lead to an over-estimation, which the AE value seems to be. On the other hand DE, ME and MA techniques are not capable of filling the same gap with any estimates, so the values presented in Table 1 correspond to the 9-months sums. Therefore, the best approximation, in our view, must be close to the two remaining LUT estimates, at 5.3 kg N_2O-N ha^{-1}. The uncertainty of this approximation cannot be obtained based on the range of the estimates, as was done for previous years, but it is unlikely it should exceed the value for the 2006, thus we assign 0.5 kg N_2O-N ha^{-1} as an uncertainty for 2007. In 2008, the annual sums produced by DE, MA and LUT_my techniques had the largest deviation from the median. The median and average of the remaining estimates (3.7 kg N_2O-N ha^{-1}) could be considered the best approximation for this year. Based on the range of ME, AE and LUT_sy an estimate of 0.3 kg N_2O-N ha^{-1} is likely to characterise the uncertainty for 2008.

To summarise, we consider the following values to be the best approximations of the true annual fluxes at our site for the years 2004–2008 (in kg N_2O-N ha^{-1}): 6.2 (±0.1), 4.3 (±0.2), 4.2 (±0.7), 5.3 (±0.5) and 3.7 (±0.3). We think that the uncertainty values, while lacking a rigorous mathematical definition, are helpful in indicating the reliability of these approximations.

It is notable that the annual sum produced by one of the simplest approaches (annual extrapolation) is very close to the best approximates in the absence of very long gaps. However, overall the most stable technique was the single-year LUT that provided the most reasonable values for all years. Estimates from daily extrapolation and moving average, both techniques primarily operating on a daily time scale, tended to be lower than other estimates. It might be expected, however, that the widening of the averaging window would help to gap-fill larger gaps. The multi-year LUT estimates were outliers in 2004 and 2008, with lower and higher values, respectively; the 2005–2007 values are closer to both single-year LUT and best approximations. From 2004 to 2008 not only the total amounts of fertilizers decreased but also the frequency of the fertilizer applications. Therefore our original assumption regarding the stationarity of the LUT-defining parameters did not hold for the whole 5-year study period. It does not, however, invalidate the LUT_my technique, considering its reasonable performance during the years 2006 and 2007 which had long gap periods.

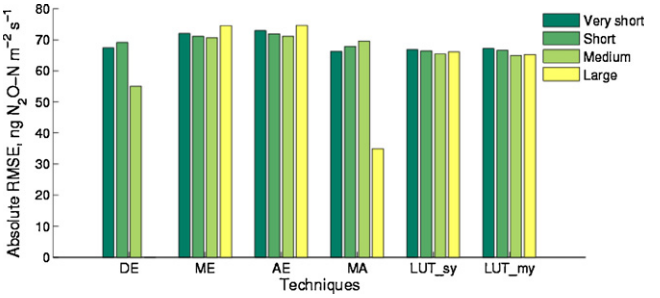

Fig. 3. Technique sensitivity to the gap length (DE—daily extrapolation, ME—monthly extrapolation, AE—annual extrapolation, MA—moving average, LUT_sy—single-year look-up table, LUT_my—multi-year look-up table).

The look-up table was the only approach that considered the environmental variables influencing the flux. This method is sensitive to the parameters selected as well as the separation limits or bin sizes. All the chosen parameters are well known as the main drivers of the N_2O emission (Dobbie and Smith, 2003a,b). The specific histories of the environmental parameters, 12-h rainfall and 5-day fertiliser applications, were based on our previous work at this site and reflected the precision of the available data. For other sites a correction to the proposed limits or even an addition of new parameters – if they can be shown to have a significant effect on N_2O flux – might be beneficial for creating better performing look-up tables. In this study the single-year LUT technique was the only method that performed well with all annual data sets (fully gap-filling the intermediate time series and producing reasonable estimates of the annual sums).

Table 2
Performance characteristics of the analysed techniques grouped by year, averaged over 50 scenarios.

Statistic	Year	DE	ME	AE	MA	LUT_sy	LUT_my
R^2	2004	0.053	0.025	0.000	0.047	0.010	0.008
	2005	0.090	0.032	0.000	0.091	0.014	0.012
	2006	0.119	0.035	0.000	0.069	0.023	0.012
	2007	0.177	0.044	0.000	0.155	0.020	0.022
	2008	0.157	0.033	0.000	0.126	0.039	0.031
Absolute RMSE, ng N_2O-N m^{-2} s^{-1}	2004	111.171	158.707	160.096	139.070	159.932	159.638
	2005	39.658	56.930	57.450	49.910	57.379	57.240
	2006	37.513	55.852	56.128	39.191	26.666	26.023
	2007	32.203	50.731	51.492	40.529	51.424	51.132
	2008	25.619	38.102	38.472	31.622	38.043	38.437
Relative RMSE	2004	0.699	0.985	0.992	0.869	0.992	0.990
	2005	0.665	0.956	0.964	0.830	0.966	0.963
	2006	0.688	0.974	0.977	0.713	0.477	0.455
	2007	0.607	0.924	0.937	0.747	0.936	0.931
	2008	0.637	0.949	0.958	0.788	0.948	0.960
Mean absolute error, ng N_2O-N m^{-2} s^{-1}	2004	56.832	85.304	85.270	71.065	86.206	84.376
	2005	18.898	30.351	30.341	24.040	30.192	29.967
	2006	21.503	34.537	34.397	21.598	7.944	7.020
	2007	17.149	28.838	28.910	21.701	29.125	29.204
	2008	14.554	23.560	23.506	17.998	23.577	24.991
Bias error, ng N_2O-N m^{-2} s^{-1}	2004	0.352	−1.695	−1.788	−0.243	−1.634	−6.085
	2005	0.018	0.088	−0.471	−0.086	−0.317	−0.033
	2006	−0.284	−0.791	−2.269	−1.193	0.834	−3.012
	2007	−0.760	−0.609	−0.517	−0.483	−0.270	0.236
	2008	−0.406	0.226	0.320	−0.844	−0.133	4.866

3.2. Statistical evaluation of techniques' performance

The results of the statistical evaluation of the techniques' performance are presented in Table 2. These figures indicate the techniques' ability to reliably predict 30-min fluxes, as opposed to the quality of the annual sums estimates discussed in the previous section. Overall, the comparison of dimensionless statistics with those of CO_2 gap-filling techniques (Moffat et al., 2007) indicates a low predictive ability of the approaches presented here. Techniques operating on a daily time scale (DE, MA) are more resistant to outliers, as can be seen from the comparison of R^2 and RMSE values by year, which is an important quality for N_2O gap-filling. On the other hand, inter-annual comparison highlights the fact that the statistical evaluation itself is sensitive to the techniques' ability to fill gaps of various lengths. The characteristics of the AE gap-filling are not entirely unexpected; for example, R^2 cannot be anything but zero for this technique.

Analysis of the technique performance when gap-filling the gaps of various length (Fig. 3) shows that some of the lower values (MA large gaps or DE medium gaps) are not the results of good gap-filling but rather a by-product of partial gap-filling of the time series, since they are associated with lower values of annual sums. Both LUT techniques performed marginally better than other techniques. These uncertainties associated with partial gap-filling could be addressed in further studies by incorporating other statistical methods or tools that would be resistant to this phenomenon. It is hard to ascertain how meaningful the statistical results are, but we hope that they will serve as benchmarks for forthcoming gap-filling techniques and will be assessed against results from other sites as well. Thus, we think that the decision regarding applicability of a particular technique should be based on other considerations, such as physical basis or model complexity. Furthermore, it should be noted that the ability of a technique to predict 30-min fluxes might not be related to the quality of prediction of annual sums, as the comparison of annual sums vs. statistical parameters demonstrates.

4. Conclusions

In this study, a number of techniques were investigated for the purpose of gap-filling a five-year time series of EC measured N_2O fluxes from a grassland ecosystem. We considered simple extrapolation methods available in the literature, as well as LUT approach adopted for N_2O gap-filling. Overall the single-year LUT technique produced the most stable results across all years and varying amount of gaps; the simpler annual extrapolation technique was also shown to produce reasonable outcomes. Techniques operating on a daily time scale, such as daily extrapolation or moving average were naturally incapable of gap-filling large or extremely large gaps encountered in our time series. As might be expected, the techniques' estimates were closer to each other when the flux time series had fewer gaps. The continuation of this study and the use of these methods under different environmental conditions are necessary to fully understand and explore agricultural emissions of N_2O.

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