

1 **Final Author Response**

2

3 We are grateful to both anonymous referees for their overwhelmingly positive comments on
4 our manuscript.

5 Referee 2 recommends “accepting the manuscript as is”. We are thrilled by this endorsement,
6 and therefore no specific responses to this referee are required.

7 Referee 1, by contrast, finds the manuscript somewhat too long and too detailed. We consider
8 this a fair opinion. When describing experimental work which consists of new combinations
9 of approaches, or contains new aspects, authors have to find a balance between, on the one
10 hand, presenting a concise and readable story, and on the other hand, giving enough detail to
11 convince referees and readers that the experimental procedures and data analysis are of high
12 standard. With this manuscript, we have certainly tended to the latter. We highly appreciate
13 the referee’s clear guidance which passages should be considered for shortening, and we state
14 below in the replies to specific comments how we intend to follow these suggestions.

15

16 **Note regarding minor changes to the data**

17 The algorithm employed by us to gap-fill the CO₂ fluxes is contained in the “OzFluxQC”
18 software provided by Peter Isaac et al. Since the submission of our discussion paper, there
19 have been a number of updates of this software package. One of these updates includes a
20 correction that causes minor alterations of the nocturnal CO₂ fluxes in our dataset, which in
21 turn leads to minor changes in the CH₄ and N₂O emission estimates. We have also, during
22 the preparation of a companion paper by Hunt et al., revisited the low-turbulence filtering
23 threshold. For consistency, we would like to use the same version of our dataset in both
24 papers. The revised filtering alters the data selection for both the GGR and NSR method
25 somewhat, and thus indirectly also alters the daily estimates of the GGR (NSR) method on
26 some days (nights). Due to these two changes in the data analysis procedure, many numbers
27 given in our Results section will be changed by a few percent. These changes are, however,
28 small enough that the conclusions are not affected; overall, our two methods and their
29 combination appear rather robust. In particular, the estimate of annual N₂O emissions will
30 stay within the uncertainty limits given in the Discussion paper.

31

32 **Response to Anonymous Referee #1**

33 **General Comments**

34 “As the authors point out in their title the two approaches are complementary: the first is
35 appropriate for the well–developed atmospheric turbulence often experienced in the daytime
36 while the second is applicable on calm nights. However, this could introduce some
37 uncertainty when there is a diurnal cycle in gas emissions as has been observed in other field
38 studies of N₂O production. Nonetheless, the paper should prove useful to other researchers of
39 the target gas emissions. The authors point out that their study was initiated because of the
40 high cost of fast sensors appropriate for eddy covariance measurements of CH₄ and N₂O,
41 most notably for N₂O.”

42 Reply: If we understand the referee’s point correctly, it is that combining two methods which
43 are always applied at different times of day, and thus not compared to each other, carries the
44 risk of misrepresenting the diurnal cycle of gas emissions. This appears to be a comment in

1 passing, rather than a criticism of our approach. We agree that in principle such
2 misrepresentation is possible; however, by applying each method only in the conditions that it
3 is best-suited for, in practice we minimise potential biases for both methods.

4 “I have some areas of concern. One is the length of the MS. It is well written, but I feel that
5 the great detail in it makes it longer and more discursive than need be. Some examples
6 follow:”

7 Reply: we are happy to shorten the manuscript in some places, see replies to specific
8 comments.

9

10 Specific Comments

11 “p.6: The sampling procedure for the FTIR spectrometer is described. It was found that
12 changes were needed to attain the desired sensitivity and a new system was used. Since the
13 first system was unsatisfactory it should be enough to cut to a short description of the second,
14 without wading through an unnecessary page of detail.”

15 Reply: The second sampling procedure achieved an improvement over the first in terms of
16 precision of the gas mole fractions, but at the cost of halving the data yield. We disagree with
17 the description of the first sampling procedure as “unsatisfactory”. The larger part of the
18 presented dataset (almost 14 months) was collected with the first procedure, and another 6
19 months with the second. Both procedures thus need to be described.

20 “p.10: I am not expert in gap-filling procedures, but I find it surprising that gap-filling was
21 apparently used so freely. Perhaps the authors could quantify just how much.”

22 Reply: We are not quite sure what exactly the words “so freely” refer to. Choice of threshold
23 value, or of threshold variable? Both are discussed in the companion manuscript by Hunt et
24 al. which will be submitted within the next month.

25 At our sites, ca. 45 % of night-time data fell below the low-turbulence threshold, which is
26 similar to many other sites around the world. We add in Section 3.3.1 a half-sentence giving
27 the fraction of nights available for the NSR method.

28 “p.10, Eq.5: Is it acceptable to use whole-night averages rather than shorter term
29 determinations in this equation and how are “sufficiently calm” nights identified?”

30 Reply: If the fluxes of CO₂ and the gas of interest were perfectly correlated in time, then the
31 length of the averaging period would not matter at all. We do not know the true fluxes, but we
32 can take the correlation coefficient between the mole-fraction gradients for guidance. The
33 lower this correlation, the poorer we expect the whole-night estimate to be, which is why we
34 excluded regressions with low R². If one tried to apply the NSR method with shorter
35 periods, then the number of points to determine the regression slope would be reduced
36 (increasing the uncertainty), and the random error of the CO₂ flux would propagate into the
37 flux estimate for the gas of interest. So, while theoretically shorter periods would allow to
38 account for out-of-sync drivers of the gas fluxes, in practice such accuracy gain is unlikely to
39 be achieved, because it would be overwhelmed by increased random error.

40 There is also a theoretical argument for high temporal correlation between the gas fluxes: the
41 soil microbial processes that produce (or consume) CO₂, CH₄ and N₂O are all soil-
42 temperature dependent and should thus co-vary.

43 The selection procedure for calm nights is described in the paragraph following the sentence
44 with “sufficiently calm” (last of Section 2.4).

1 “pp. 11 & 12: To me, the description of the Soil and Vegetation conditions and CO₂ fluxes
2 contains more detail than is needed and could be shortened.”

3 Reply: Yes we are happy to make some cuts here.

4 “p.13: Define non-resolvable gradients. My guess is that they exhibit a change from negative
5 to positive (or vice versa) within the gradient”

6 Reply: These are defined in the preceding sentence. We’ll link the two sentences up by
7 inserting the word “such” (“Such non-resolvable mole-fraction differences…”).

8 “pp. 15-17: These pages, which discuss various aspects of the GGR method, are good
9 examples of the highly detailed patches in the MS that I think could be shortened.”

10 Reply: Section 3.2.3 is already short, and we would agree to cut another sentence or two.
11 Section 3.2.4. is also relatively short and we use it to show that the footprint of the GGR
12 method contrasts with that of the NSR method described in a later section. Section 3.2.5 will
13 be scrutinised for possible cuts.

14 “pp. 18-19 (section 3.3.3 on footprints and nocturnal fluxes): long discussion”

15 Reply: We agree that some minor cuts are possible, but the substance of this section should
16 stay, because it shows that the footprint requirements can be a major caveat of the NSR
17 method.

18 “I have some concern about the use of the term turbulent diffusivity, as in Eq. 1. My
19 understanding is that it should be used in a partial derivative equation rather than a finite
20 difference one like Eq. 1 so that

21 $F_{\chi} = -K \partial\chi / \partial z$,

22 as, for instance, in Thom (1975, Momentum, Mass and Heat Exchange of Plant Communities
23 in “Vegetation and the Atmosphere” Vol.1, Ed. J.L Monteith, pp.57-109, Academic Press,
24 London). This allows for a non-linear form for $\chi(z)$ and a non-steady state. I prefer to
25 describe K as used by the authors in Eq.1 as a transport or transfer coefficient. I haven’t gone
26 through the ramifications that might arise from using Eq.1 in the present context. It may be
27 that in the end, both Eq.1 and the partial differential equation above give the same answer.”

28 Reply: The referee is correct that the vertical diffusivity is defined using the partial derivative
29 with height. Our Eq. 1 is already simplified for the practical application, where measurements
30 can only be taken at discrete heights to approximate $\partial[\chi] / \partial z$. We will insert a sentence just
31 above the equation to clarify this. There are no further ramifications of our early
32 approximation. If we wrote the partial derivatives, for $[\chi]$ in Eq. 1 and for $[\text{CO}_2]$ in Eq. 3, then
33 the ratio of these two would appear in Eq. 4, and the simplification to finite differences would
34 have to be employed at that stage, with identical result. Technically this is a linearisation;
35 however, Eq. 4 is accurate not only if the profile shapes are linear with height, but also if the
36 profiles shapes deviate from linearity in the same fashion for both gases – which is what
37 similarity theory would predict under the assumption that the sources/sinks for both gases
38 were co-located.

39 Regarding the name of K, our simplification does not change the concept or the units, so it is
40 still correct to call it “diffusivity”. By contrast, the expression “transfer coefficient” is
41 ambiguous. Stull (1988, An Introduction to Boundary-Layer Meteorology) uses it
42 synonymous with diffusivity. Phillips et al. (2007, see manuscript references) use it for
43 $K / \Delta z$. In other contexts, it is a dimensionless number (e.g. bulk transfer coefficient). We
44 prefer to avoid such ambiguity.

1 “pp.23-25: These discussion pages make good points, particularly the opening paragraph on
2 p.25 which recommends combining the GGR and NSR techniques to give long term means
3 since the GGR method yields more data during the day than at night and the NSR method is
4 nocturnal only. The authors point out that their combination optimises data usage.”

5 Reply: Thank you for this affirmative comment. No changes required here.

6

7 Technical Comments

8 “I noticed some typos in the manuscript:

9 p.7, line 4: thermostate for thermostatsat”

10 Reply: “thermostat” is the correct spelling, this will be fixed.

11 “p.7, line26: instationary for non-stationary”

12 Reply: “instationary” is a correct and common technical term in fluid dynamics, no need to
13 change.

14 “p.28, line 17: contents for content”

15 Reply: This will be corrected.

16

17

Combining two complementary micrometeorological methods to measure CH₄ and N₂O fluxes over pasture

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Abstract

New Zealand's largest industrial sector is pastoral agriculture, giving rise to a large fraction of the country's emissions of methane (CH₄) and nitrous oxide (N₂O). We designed a system to continuously measure CH₄ and N₂O fluxes at the field scale on two adjacent pastures that differed with respect to management. At the core of this system was a closed-cell Fourier-transform infrared spectrometer (FTIR), measuring the mole fractions of CH₄, N₂O and carbon dioxide (CO₂) at two heights at each site. In parallel, CO₂ fluxes were measured using eddy-covariance instrumentation. We applied two different micrometeorological ratio methods to infer the CH₄ and N₂O fluxes from their respective mole fractions and the CO₂ fluxes. The first is a variant of the flux-gradient method, where it is assumed that the turbulent diffusivities of CH₄ and N₂O equal that of CO₂. This method was reliable when the CO₂ mole-fraction difference between heights was at least 4 times greater than the FTIR's resolution of differences. For the second method, the temporal increases of mole fractions in the stable nocturnal boundary layer, which are correlated for concurrently-emitted gases, are used to infer the unknown fluxes of CH₄ and N₂O from the known flux of CO₂. This method was sensitive to "contamination" from trace gas sources other than the pasture of interest and therefore required careful filtering. With both methods combined, estimates of mean daily CH₄ and N₂O fluxes were obtained for ~~60~~56 % of days at one site and ~~77~~73 % at the other. Both methods indicated both sites as net sources of CH₄ and N₂O. Mean emission rates for

1 one year at the unfertilised, winter-grazed site were 8.29 (± 0.9479) nmol CH₄ m⁻² s⁻¹ and
2 0.4038 (± 0.018) nmol N₂O m⁻² s⁻¹. During the same year, mean emission rates at the
3 irrigated, fertilised and rotationally-grazed site were 7.8.9 ($\pm 0.89.79$) nmol CH₄ m⁻² s⁻¹
4 and 0.5758 (± 0.019020) nmol N₂O m⁻² s⁻¹. At this site, the N₂O emissions amounted to
5 1.1921 (± 0.15) % of the nitrogen inputs from animal excreta and fertiliser application.

6 7 8 **1 Introduction**

9 The accurate assessment of greenhouse gas (GHG) fluxes between the biosphere and the
10 atmosphere is crucial to understand driving mechanisms of global climate change. While net
11 ecosystem exchange (NEE) of carbon dioxide (CO₂) fluxes are being measured for multiple
12 years at over 400 sites around the globe, using the eddy-covariance method (Baldocchi,
13 2014), continuous methane (CH₄) and nitrous oxide (N₂O) measurements are still comparably
14 sparse at the ecosystem scale (Nicolini et al., 2013). However, the accounting of full GHG
15 budgets is especially important for agroecosystems as they are the largest global source of
16 N₂O and CH₄ emissions (Montzka et al., 2011). For instance, Leahy et al. (2004) showed that
17 N₂O and CH₄ emissions on managed grasslands have the potential to fully counteract the CO₂
18 sink strength in these ecosystems. This has been confirmed by a European wide synthesis
19 study done at 10 different grasslands sites over two years (Soussana et al., 2007). Moreover,
20 among soils from different land-use types, those from grasslands have been shown to have the
21 highest rates of N₂O emissions, due to high microbial activity stimulated by high soil C and N
22 content (Schaufler et al., 2010).

23 In order to assess effects of management and land-use changes on net GHG budgets, methods
24 are required to measure GHG fluxes at the scale at which agroecosystems are managed, which
25 is the field scale. Long-term field-scale studies of GHG fluxes in New Zealand's pastoral
26 agroecosystems are so far restricted to CO₂ only (Nieveen et al., 2005; Mudge et al., 2011).
27 Yet, 48 % of the country's total GHG emissions are CH₄ and N₂O emissions from agriculture
28 (MfE, 2015). We therefore began a project to simultaneously measure the exchange rates of
29 all three GHGs from pastures on a commercial dairy farm. In this paper, we describe and
30 critically assess our methods to measure CH₄ and N₂O fluxes and report results for these from
31 the first 20 months of this project. The reporting of CO₂ fluxes is kept brief here; for detailed

1 derivation and discussion see Hunt et al. (2015). ~~The same data also contribute to the~~
2 ~~synthesis study by Voté et al. (2015(2016, this issue).~~

3 Particularly suitable for the field scale are micrometeorological methods (Denmead, 2008). Of
4 these, the eddy-covariance method has steadily increased in popularity since new types of fast
5 and precise gas analysers for CH₄ and N₂O were shown to be suitable (Eugster et al., 2007;
6 Kroon et al., 2007; Tuzson et al., 2010). In managed grasslands, eddy-covariance
7 measurements with such instruments have since been undertaken e.g. by Neftel et al. (2010),
8 Merbold et al. (2014), Hörtnagl and Wohlfahrt (2014), and Schrier-Uijl et al. (2014).

9 Disadvantages of the eddy-covariance method are that the fast analysers for CH₄ and N₂O are
10 expensive, often specific to a single gas, and frequently associated with large measurement
11 errors during periods of small fluxes (Kroon et al., 2010). Therefore, we pursue an alternative
12 approach: we use a slow, closed-cell, multi-gas analyser (FTIR spectrometer, see Section 2.1
13 for details), and we combine two other micrometeorological methods in order to maximise the
14 GHG flux information that can be gained from such measurements. One of these methods is a
15 variant of the flux-gradient method (Denmead ~~et al.~~, 2008) which we shall denote as the gas-
16 gradient ratio (GGR) method. It relies on the equality of turbulent diffusivities for different
17 gas species and is fully described in Section 2.3. The other method was first applied by
18 Kelliher et al. (2002) and will here be referred to as the nocturnal storage-ratio (NSR) method.
19 It exploits that during calm nights with stable surface-layer stratification, gases emitted at the
20 surface accumulate over time in the surface layer, much like in a natural “big chamber”. For
21 gases originating from the same locations, their mole-fraction increases in the surface layer
22 are strongly correlated, and they are easily detectable (see Section 2.4).

23 Both methods are essentially tracer-ratio methods, where we use CO₂ as the tracer, or
24 reference gas. Therefore, concurrent measurements of CO₂ fluxes are required, for which we
25 employ standard eddy-covariance instrumentation. For calm nights, we follow routine
26 practice to discard measured CO₂ fluxes (considering them unreliable) and replace them with
27 modelled values from a gap-filling algorithm. The suitability of these modelled tracer fluxes
28 for the NSR method is assessed as part of our data analysis.

29 Our choice of instruments and methods was largely guided by practical considerations.
30 Firstly, we aimed to undertake a paired-site study on two neighbouring pastures with different
31 managements. Rather than having to acquire and maintain four gas analysers, as required for
32 the eddy-covariance method with two gases at two sites (in addition to the standard CO₂-flux

1 instrumentation), it appeared a sensible alternative to employ a single multi-gas instrument
2 with a switching system, fed via long air-intake lines. Secondly, one of the two sites was
3 irrigated by a pivot irrigator that regularly passed over with a clearance height of ca. 2.5 m.
4 This constraint on measurement height made it undesirable to operate bulky open-path
5 analysers. While these conditions were very specific to our experiment, we believe that our
6 combination of methods may be useful and attractive in other measurement situations, too.

7 It is not intended here to give full greenhouse gas budgets of the dairying operations. That
8 will be the subject of future research, based on longer time series of flux measurements. In
9 particular, this study does not consider CH₄ emissions from grazing animals. We exclude all
10 periods from our analysis during which cattle were present. Following standard practice of
11 dairy farms in New Zealand, these grazing events were of only 1 to 2 d duration each, at an
12 animal density of order 100 head ha⁻¹. At this density, the fluxes of CH₄ and CO₂ would be
13 dominated by eructation and respiration, respectively, from the cattle. The cattle were then
14 moving point sources in a confined space; to measure accurate and representative fluxes from
15 these would be a challenging task, for which a number of micrometeorological methods have
16 been developed and tested elsewhere (Laubach et al., 2008; [Laubach et al., 2013](#); Felber et
17 [al., 2015](#)). Instead, we aim to determine exchange rates of the pasture surface itself, which for
18 CH₄ are expected to be at least two magnitudes smaller than the emission rates from a grazing
19 cattle herd.

20 The first objectives of this paper are to describe the GGR and NSR method in detail and to
21 identify for each the conditions in which it reliably operates (or otherwise). Based on that
22 assessment, we aim to specify data-filtering criteria for each method. We then compare results
23 between the two methods and assess how they should be combined to obtain reliable annual
24 mean fluxes of CH₄ and N₂O from the pasture surfaces. In the case of the irrigated pasture, we
25 are able to relate the measured N₂O fluxes to the nitrogen (N) inputs from fertilisation and
26 excreta deposition.

27

28

1 2 Materials and methods

2 2.1 Site and farming operations

3 The study was conducted on an intensively managed dairy farm (ca. 900 cows on 382 ha) on
4 the Canterbury Plains, South Island, New Zealand (Lat $-43^{\circ} 35' 30.6''$, Lon $171^{\circ} 55' 36.6''$;
5 [208204](#) m a.s.l.). The soil is a Lismore silty loam which is moderately stony (~~clay 15–25 %~~)
6 and well-drained. The farm was converted to irrigated dairying in 2008. The irrigated pasture
7 is dominated by ryegrass (*Lolium perenne* L.) with a minor fraction of white clover
8 (*Trifolium repens* L.). The main irrigated area (263 ha) is circular and subdivided into 19
9 similar-sized paddocks. Part of the farm is an area of non-irrigated land, wedged between this
10 farm's irrigated circle to the S and a similar irrigated circle of the neighbouring farm to the W
11 (Fig. 1). We established two measurement sites in mid-2012, one inside the irrigated circle at
12 ca. 140 m distance from the perimeter, the other inside the dryland wedge, at a similar
13 distance from its boundaries to S and W. Instrumentation connected to both sites was operated
14 in a hut placed at the boundary between the irrigated and the dryland pasture (Fig. 1).

15 The farm managers monitored volumetric water ~~content~~content (VWC) of the pasture soil
16 (10 to 40 cm depth) and used the pivot irrigator to keep VWC above $0.2 \text{ m}^3 \text{ m}^{-3}$. The irrigated
17 pasture was fertilised 9 times annually, on the majority of occasions with urea; in the 2012/13
18 season, the total application rate was $18.3 \text{ g N m}^{-2} \text{ yr}^{-1}$. Throughout the milking season
19 (September to late April) each paddock was grazed 8 to 12 times, usually by 400 to 500 cows.
20 For our measurement site, times of grazing events were recorded. Effluent collected at the
21 milking shed was recycled to the pasture via irrigator attachments. We will refer to our
22 measurement site on the irrigated, fertilised, rotationally-grazed pasture as the “IFR site~~”~~;
23 for future reference in the Fluxnet network it has been named “NZ-BFm” (D. Papale, pers.
24 comm., 2016).

25 The dryland pasture was used for winter-grazing once in July 2012, prior to the start of our
26 measurements, and once in May 2013, otherwise it was not managed during the 2012/13
27 season. The measurement site there is labelled as the “UW site” (unirrigated, unfertilised,
28 ~~winter-grazed~~); its future Fluxnet name will be “NZ-BFu”. On 28 October 2013 the pasture
29 was sprayed with herbicide, followed by sowing kale (*Brassica oleracea* L., cultivar “corka”)
30 on 20 November, to provide a forage crop for winter 2014.

31

1 2.2 Instrumentation

2 2.2.1 Mole fraction measurements of CH₄, N₂O and CO₂

3 Mole fractions of the trace gases CO₂, CH₄, N₂O and CO were measured simultaneously with
4 a Fourier-transform infrared (FTIR) trace gas analyser built at the University of Wollongong
5 (Griffith et al., 2012). The analyser is equivalent to the now commercially available
6 Spectronus analyser (Ecotech, Knoxfield, VIC, Australia). This instrument performs
7 broadband-spectrum absorption measurements and uses an optimisation algorithm called
8 MALT to retrieve mole fractions of the trace gas species of interest from the measured
9 infrared spectra. For details of both the FTIR hardware and the MALT algorithm see Griffith
10 et al. (2012).

11 The FTIR was housed in the hut midway between the two measurement sites. From two
12 heights ($z_1 = 0.76$ m, $z_2 = 1.96$ m), at each site, air was drawn continuously via 170 m long
13 nylon tubing, buried at 0.2 m depth, into 10 L stainless steel cylinders (ballast tanks) inside
14 the hut. A fifth air intake was at 10 m height, next to the hut, to sample gas mole fractions
15 representative of the wider surroundings. All five air streams were drawn in parallel with a
16 dual-head diaphragm pump (2107AC, Gardner Denver Thomas, Sheboygan, Wisconsin,
17 USA). The FTIR was operated in discrete cell-fill mode, sampling air from each of the five
18 ballast tanks once over the course of a 30-min cycle. During every 29th cycle, the
19 measurement from 10 m was skipped, and instead a sample was taken from a cylinder
20 containing air with known mole fractions of the gases of interest (“target tank”), to check for
21 calibration drifts. An external manifold unit with solenoid valves switched between the
22 different sample and calibration intakes. A four-diaphragm oil-free vacuum pump (MV 2 NT,
23 Vacuubrand, Wertheim, Germany) served to evacuate the FTIR’s measurement cell to about
24 2.5 hPa and then to refill it with sample air. Before entering the cell, the sample air was dried
25 in two stages, with a Nafion[®] drier and a magnesium perchlorate trap. This resulted in water
26 vapour mole fractions $< 10 \mu\text{mol mol}^{-1}$, which were included in the mole fractions retrieved
27 by the MALT algorithm.

28 The FTIR was run in static mode according to the following measurement cycle procedure:
29 1) cell and manifold evacuation for 120 s, reaching ca. 3 hPa; 2) cell fill to 900 hPa (for up to
30 a maximum duration of 120 s); 3) cell pressure stabilisation for 60 s, 4) spectrum collection
31 and analysis for 60 s, then wait to the end of 6 min and repetition of these steps for the next

1 intake line. Because the previous sample was not completely removed from the cell, the
2 measured mole fractions were corrected for the residual sample in the cell from the previous
3 measurement. Since $[\text{N}_2\text{O}]$ and $[\text{CH}_4]$ differences between intakes were often less than the
4 resolution limits (see Results section), we tested whether the measurement precision could be
5 improved by allowing more time for Step 4). This was found to be the case, therefore in a new
6 measurement cycle, the time for Step 4) was increased 7-fold, allowing far more individual
7 spectra to be collected and analysed for each cell fill. As the new cycle required 60 min total
8 duration for the five intakes, intakes at only one site could be sampled each half-hour.
9 Therefore, the increased precision came at the cost of halving the data yield. With more time
10 per intake available, the cell-fill pressure was increased to 950 hPa (to achieve a further
11 modest gain in precision) and up to 140 s were allowed for the filling process (Step 2). The
12 new cycle was used from 15 October 2013 onwards.

13 The hut temperature was controlled at $20 (\pm 2) ^\circ\text{C}$, and the measurement cell of the FTIR was
14 thermostat-regulated to $30 ^\circ\text{C}$. Cell temperature during measurements was recorded and used
15 in the retrieval of gas mole fractions from the spectra (Griffith et al., 2012). The cell
16 temperature was found to respond to changes in hut temperature at a rate of 0.01 K K^{-1} .

17 The FTIR was calibrated with three different gas cylinders of clean southern-hemisphere
18 background air, spiked with varying mole fractions for each species of interest. These
19 cylinders had been filled and their contents composition measured by the National Institute of
20 Water & Atmospheric Research (Wellington, New Zealand), following the standards of the
21 WMO's Global Atmospheric Watch programme. Calibrations were carried out 8 times over a
22 20-month period and yielded very consistent results: the calibration slopes (of measured vs
23 nominal mole fractions) showed no systematic variation over time, and their relative standard
24 deviations were 0.14 % for CO_2 , 0.08 % for CH_4 , and 0.16 % for N_2O .

25

26 2.2.2 CO_2 flux measurements and gap-filling

27 At each site, an eddy covariance (EC) system was operated to measure the fluxes of CO_2 ,
28 water vapour, heat and momentum at 1.86 m above ground. The system consisted of an
29 enclosed infrared gas analyser to measure CO_2 and H_2O vapour concentrations (LI-7200, LI-
30 COR Biosciences, Lincoln, NE, USA) and an ultrasonic anemometer to measure the wind
31 vector and temperature (WindMaster Pro, Gill Instruments, Lymington, Hampshire, UK). The

1 raw 20 Hz time series were collected with an Analyzer Interface Unit (LI-7550, LI-COR
2 Biosciences, Lincoln, NE, USA). Data corrections, averaging and flux computation were
3 performed with the open-source software EddyPro (LI-COR Biosciences, Lincoln, NE, USA).
4 Further details of the EC setup are given in Hunt et al. (~~2015~~[\(2016, this issue\)](#)).
5 The CO₂ flux data were filtered to exclude grazing periods, periods of instationary wind
6 conditions, and periods of low turbulence. The last were flagged by $\sigma_w = 0.4012$ m s⁻¹, where
7 σ_w is the standard deviation of vertical wind speed, following Acevedo et al. (2009) who argue
8 this is a more selective filter than the commonly-used friction velocity threshold. The ensuing
9 data gaps were filled with an artificial-neural-network method, known as the SOLO (self-
10 organizing linear output) model (Hsu et al., 2002), which has been shown to be accurate on a
11 daily basis (Eamus et al., 2013).
12 At the IFR site, small-scale CO₂ flux measurements with four automated respiration chambers
13 (LI-8100A, LI-COR Biosciences, Lincoln, NE, USA) were available for part of the
14 measurement period. These chambers were placed ca. 3 m from the EC mast, within a fenced
15 area from which the cows were excluded. They received the same irrigation and fertilisation
16 applications as the surrounding paddock. The grass inside the chambers was cut manually
17 when the surrounding paddock was grazed, and on these occasions, urea was hand-applied to
18 simulate the additional N input from excreta deposited by grazing animals. Data from these
19 chambers are used to corroborate the nocturnal CO₂ fluxes obtained from EC measurements
20 and SOLO gap-filling.

21

22 2.2.3 Ancillary measurements

23 Alongside the EC measurements, each site was equipped to record half-hourly averages of
24 precipitation, radiation, wind speed and direction, PAR, relative humidity, air temperature,
25 soil heat flux and direct and diffuse radiation. Also, soil temperature and soil VWC were
26 recorded in three separate profiles per site, at depths of 5, 10, 25, and 50 cm. Soil temperature
27 was measured with copper/constantan thermocouples and VWC with time-domain
28 reflectometry probes (SM300, Delta-T Devices, Burwell, Cambridge, UK).

29

1 **2.3 Gas-gradient ratio method** **Sentence before Eq. 1 added in response to Ref. 1**

2 The gas-gradient ratio (GGR) method is a variant of what is commonly known as the flux-
3 gradient method. In the latter, the flux F_χ of a gas species χ is computed as the product of the
4 (negative) vertical concentration gradient with a turbulent diffusivity. In practice, the
5 infinitesimal gradient is approximated by finite differences, which gives:

$$6 \quad F_\chi = -K_\chi C_{\text{air}} \frac{[\chi]_1 - [\chi]_2}{z_1 - z_2} \quad (1)$$

7 where $[\chi]$ is the mole fraction and K_χ the turbulent diffusivity of χ , C_{air} the molar density of
8 dry air, z height above ground, and the subscripts 1 and 2 indicate measurement heights.
9 Upward fluxes (away from the surface) are represented with positive values, following the
10 micrometeorological convention. The diffusivity is parameterised using similarity theory
11 (Oke, 1987; Denmead, 2008). Often, the parameterisation is based on momentum exchange;
12 this approach is also known as the aerodynamic method (Oke, 1987; Denmead, 2008). There,
13 the diffusivity of momentum, K_m , is usually specified as:

$$14 \quad K_m = k u_* z \phi^{-1} \quad (2)$$

15 where k is the von-Kármán constant, u_* friction velocity, and ϕ a function of stratification. To
16 make use of (2), the ratio of the diffusivities for momentum and mass, K_m/K_χ (named the
17 turbulent Schmidt number) must be known; yet, how this number varies with height scale and
18 flow statistics in the surface layer is difficult to measure and different approaches have
19 yielded contradictory results (Wilson, 2013). By contrast, there is less ambiguity about the
20 relationships between the diffusivities of different scalar variables, such as heat, moisture and
21 trace gas concentrations. For a pair of scalars with similar spatial distribution of their
22 source/sink locations, the diffusivities are equal, except for small differences between heat
23 and gases with regard to effects of thermal stratification. Thus, by measuring the flux and the
24 gradient of a reference scalar variable, the diffusivity can be obtained. For example, Phillips et
25 al. (2007) used heat as the reference variable to derive N_2O fluxes over pasture, and Griffith et
26 al. (2002) used water vapour as the reference to obtain fluxes of CO_2 , CH_4 and N_2O . In both
27 cases, this method was largely restricted to daytime (because fluxes and gradients of heat and
28 water vapour are small at night). Here, we use CO_2 as the reference variable, a choice
29 successfully employed to measure CH_4 emissions from rice crops (Miyata et al., 2000,
30 McMillan et al., 2007). From Eq. (1), its turbulent diffusivity is given by:

$$K_{\text{CO}_2} = -\frac{F_{\text{CO}_2}}{C_{\text{air}} \Delta[\text{CO}_2]} \Delta z \quad (3)$$

where Δ indicates the difference between heights, i.e. $\Delta[\text{CO}_2] = [\text{CO}_2]_1 - [\text{CO}_2]_2$ and $\Delta z = z_1 - z_2$. Assuming $K_\chi = K_{\text{CO}_2}$ where χ now refers to the non- CO_2 gas species, and inserting Eq. (3) back into Eq. (1) results in:

$$F_\chi = \frac{\Delta[\chi]}{\Delta[\text{CO}_2]} F_{\text{CO}_2} \quad (4)$$

The gas-gradient ratio, $\Delta[\chi]/\Delta[\text{CO}_2]$, is independent of flow properties, such as wind speed or stratification.

Since fluxes and gradients of CO_2 undergo sign changes in the morning and evening, periods including these transitions will need to be excluded. It is expected that these periods of near-zero $[\text{CO}_2]$ gradients often coincide with periods that are unsuitable for the GGR method anyway, because the surface-layer undergoes transitions between unstable and stable stratification and flow conditions will not be steady enough to define a meaningful diffusivity.

2.4 Nocturnal storage-ratio method

During clear night-time periods, cooling of the ground surface due to long-wave radiation losses causes stable stratification of the atmospheric surface layer. In this nocturnal inversion layer, turbulent exchange is suppressed and wind speeds are low. Consequently, trace gases emitted from the ground, or the biosphere near the ground, accumulate in this layer over time. This accumulation process underlies the principle of the nocturnal storage-ratio (NSR) method. Provided that the spatial distribution and temporal pattern of emissions are similar for different gas species, their mole-fraction increases over time must be strongly correlated with each other. This is illustrated in Fig. 2, where changes in $[\text{CH}_4]$, $[\text{CO}_2]$ and $[\text{N}_2\text{O}]$ track each other well: the mole fractions increase sharply when σ_w falls below 0.12 m s^{-1} , and they return towards their baseline values when σ_w rises persistently above 0.12 m s^{-1} . The correlations between the mole fractions of the trace gases are exploited to link the unknown flux of a gas species χ with the known flux of another gas, here CO_2 , as follows (Kelliher et al., 2002; Pendall et al., 2010). The relationship between the mole-fraction increases, $d[\chi]$ and

1 $d[\text{CO}_2]$, is assumed linear and expressed by the slope of a linear regression. The flux of χ is
2 then computed as the product of the CO_2 flux with the regression slope:

$$3 \quad \overline{F_\chi} = \frac{\overline{d[\chi]}}{\overline{d[\text{CO}_2]}} \overline{F_{\text{CO}_2}} \quad (5)$$

4 where the overbars indicate whole-night averages. This method is only applied for sufficiently
5 calm nights, and the regression slope determined separately for each of these.

6 During calm conditions, flux measurements by eddy covariance are notoriously unreliable.
7 Kelliher et al. (2002) and Pendall et al. (2010) used respiration chambers to measure the CO_2
8 flux instead. Here, we try a different approach. We define calm conditions by the same low-
9 turbulence threshold that is used to filter the CO_2 fluxes measured by eddy covariance prior to
10 gap-filling, as illustrated in Fig. 2. We then use the CO_2 fluxes constructed by the gap-filling
11 algorithm as inputs for the NSR method. Since Eq. (2) applies to whole nights, it is not the
12 accuracy of the half-hourly gap-filled fluxes that matters, but only the accuracy of their
13 whole-night average. We will assess the suitability of gap-filled CO_2 fluxes for deriving other
14 trace gas fluxes with the NSR method (Section 3.3.2).

15

16 **2.5 Measurement period**

17 We analyse measurements from the period 17 August 2012 to 31 March 2014, including one
18 full grazing/irrigation season, the larger fraction of a second one, and the winter between
19 these. The FTIR operated continuously, except for short maintenance and calibration periods
20 (a few hours each) and one fortnight of mains-power outage (10 to 24 September 2013)
21 following widespread windstorm damage in the region. Data collected during grazing events
22 were excluded because the CO_2 and CH_4 gradients were dominated by emissions from the
23 animals, which vary erratically with animal positions, and fluxes originating at the pasture
24 surface cannot be retrieved. At the IFR site, grazing events usually lasted only 1 to 2 d; at the
25 U UW site, there were only two grazing periods, in May 2013 and in late October 2013 (just
26 prior to the conversion to kale). The grazing dates of other irrigated paddocks, outside the one
27 containing our measurement site, were not recorded. Data filtering criteria for the GGR and
28 NSR method are explored in the Results section.

1

2

3 3 Results

4 3.1 Soil and vegetation conditions, and CO₂ fluxes Cuts in response to Ref. 1

5 Soil temperatures at 5 cm depth ranged from 2.5 °C to 22.3 °C at the IFR site and from 1.5 °C
6 to 23.5 °C at the U UW site, with the minima occurring in early July and the maxima in late
7 January. Rainfall in the year starting 17 August 2012 was 1014 mm, of which 407 mm fell in
8 the warmer half (October – March). At the IFR site, this was supplemented with 425 mm of
9 irrigation to keep the pasture well-watered throughout the grazing season. The irrigator
10 rotated continuously from mid-spring (October) to mid-autumn (early April), with a return
11 period of about 3 d, applying about 10 L m⁻². ~~The passage of the irrigator at our measurement~~
12 ~~site lasted only a few minutes, so the water application resembled a short, very intense rain~~
13 ~~shower.~~ As a result, soil VWC at 5 cm depth stayed above 0.24 m³ m⁻³. Meanwhile, the U UW
14 site experienced drought conditions from mid-December 2012 until mid-March 2013, with
15 VWC in the range 0.06 to 0.15 m³ m⁻³.

16 Over the remaining 7.5 months (after 16 August 2013), 598 mm of natural precipitation
17 occurred. In this period, the start of the irrigation was delayed until mid-November 2013, due
18 to storm damage to the irrigator. When irrigation began, VWC at 5 cm had already fallen to
19 ca. 0.15 m³ m⁻³, at both sites. This irrigation season was terminated on 19 Feb 2014 with the
20 onset of sufficient natural rainfall; the total irrigation in 2013/14 amounted to 345 mm.

21 Fig. 3 shows the time courses of daily mean CO₂ fluxes (net ecosystem exchange), computed
22 from gap-filled half-hourly fluxes, excluding days on which grazing occurred. At the IFR site,
23 each grazing event turned the pasture abruptly from a CO₂ sink (negative mean flux) to a CO₂
24 source (positive mean flux), while the leaf area index was reduced from 2.7 to 0.6 m² m⁻²
25 (Hunt et al. ~~2015., 2016~~). After a few days of regrowth, the flux reversed sign and the CO₂
26 sink strength increased steadily, until the next grazing. As a consequence of the regular
27 fertilisation and irrigation, the pasture was highly productive ~~and periods as well as amounts~~
28 ~~of CO₂ uptake were well in excess of periods and amounts of CO₂ release, except in May after~~
29 ~~the final grazing of the season. The annual net uptake of CO₂ for 2012/13 was~~
30 ~~438 (±60) g C m⁻² (Hunt et al. 2015).~~

1 The U UW site, by contrast, acted as a CO₂ sink mainly in the two spring seasons (Sept to
2 Nov) and during the growth of the kale crop in the 2013/14 summer. A prominent feature in
3 Fig. 3 is the change from sink to source in December 2012 as a consequence of the drought
4 conditions. From then until the next spring, the U UW pasture remained a weak CO₂ source.
5 ~~Over the 2012/13 year, there were non-significant net emissions of 3 (±36) g C m⁻² (Hunt et~~
6 ~~al. 2015).~~ Also clearly visible in Fig. 3 is the disturbing effect of the conversion to kale,
7 making the site temporarily a stronger CO₂ source in Nov-Dec 2013, until the kale crop had
8 established itself.

9

10 **3.2 GGR method**

11 3.2.1 Mole-fraction gradients and instrument precision

12 The short-term repeatability of the FTIR's mole-fraction measurements was determined by
13 repeatedly filling the cell with target-tank air and otherwise following the same switching-
14 cycle sequence as described in Section 2.2.1. With the original cycle (total time of 6 min per
15 intake), the standard deviation (SD) of the target-tank [N₂O] was 0.28 ppb; with the modified
16 cycle (12 min per intake), this was halved to 0.14 ppb (Table 1). For [CH₄], the SD of the
17 same target-tank samples was 0.58 ppb with the original cycle and reduced to 0.20 ppb with
18 the modified cycle. For [CO₂], the effect of the change in cycle was less pronounced, with an
19 SD of 0.15 ppm with the original cycle and 0.12 ppm with the modified cycle.

20 In Table 1, the mean mole fractions using the original cycle and the modified cycle tend to
21 differ by more than the observed SD values (particularly strongly for CH₄). The likely cause
22 of this is that with the original cycle the spectral measurements were made before the cell
23 contents had thermally equilibrated. While this lack of equilibrium causes small biases in the
24 mole fractions, the repeatability of these is not affected (as attested by the observed SD
25 values), and mole-fraction differences between intakes (GGR method) or between successive
26 nighttime hours (NSR method) are unbiased.

27 To define a „precision“, the acceptable probability of a single measurement deviating
28 randomly by more than this precision from the mean value must be specified. If we take the
29 acceptable probability as 5 %, then the precision is given as 3 times the SD of repeated
30 sampling. The resolution of a gradient measurement (mole-fraction difference between two

1 intakes) follows directly as $2^{1/2}$ times that precision: differences smaller than that cannot be
2 | considered significantly different from zero. ~~Non~~Such non-resolvable mole-fraction
3 differences between intakes occurred frequently, for all three gases. According to Eq. (3), a
4 non-resolvable gradient of gas species χ (N_2O or CH_4) simply implies a non-resolvable flux of
5 that species, in proportion. When many repeated measurements are made over time to
6 determine a mean flux, those runs with non-resolvable fluxes still make valid contributions: as
7 the standard error of the mean (SEM) decreases with increasing number of samples, the sign
8 (and magnitude) of the mean flux becomes better-resolved. Hence, small gradients of $[\text{N}_2\text{O}]$
9 or $[\text{CH}_4]$ were not removed from the dataset.

10 By contrast, $\Delta[\text{CO}_2]$ appears in the denominator of Eq. (3), with the consequence that small
11 values of it lead to highly uncertain fluxes of χ . Thus, the data must be filtered for a minimum
12 $\Delta[\text{CO}_2]$. We test this by its effect on the diffusivity, as follows.

13

14 3.2.2 Turbulent diffusivities

15 Half-hourly values for the gas diffusivity, K_{CO_2} , computed with (3), are compared in Fig. 4
16 against the momentum diffusivity, K_m , for neutral stratification, i.e. using (2) with $\phi = 1$.
17 These data are shown both for all available runs and for two selections with a minimum
18 $\Delta[\text{CO}_2]$, of 0.6 ppm and 2.4 ppm (ca. 1 and 4 times the resolution of the FTIR for CO_2). The
19 unfiltered gas diffusivity data are widely scattered, even outside the margins of the figure. The
20 smaller $\Delta[\text{CO}_2]$ threshold constrains the K_{CO_2} values, by and large, to the range -0.6 to
21 $1 \text{ m}^2 \text{ s}^{-1}$, proving that a large part of the scatter originates from small values in the
22 denominator of (3) subject to large relative errors. The larger $\Delta[\text{CO}_2]$ threshold reduces the
23 K_{CO_2} range further, to about -0.1 to $0.4 \text{ m}^2 \text{ s}^{-1}$. The positive side of this range is similar to the
24 range of momentum diffusivities and therefore considered realistic. The negative side is due
25 to sign mismatches between CO_2 fluxes and gradients. Most of these occur around the
26 morning and evening transitions, indicating that at such times the surface fluxes and surface-
27 layer gradients are not in equilibrium, and that the application of a simple flux-gradient
28 concept is then not appropriate. All runs with negative K_{CO_2} are thus excluded from further
29 analysis, in addition to the runs with $\Delta[\text{CO}_2] < 2.4$ ppm.

1 Turbulent diffusivities are positively correlated with wind speed; for K_m this is evident from
2 the dependence on u_* in (2). In Fig. 4, it is apparent that filtering with the larger $\Delta[\text{CO}_2]$
3 threshold removes almost all values measured at higher wind speeds. The higher the wind
4 speed, the better the turbulent mixing, and consequently, the smaller the vertical gradients of
5 any scalar variables. As these gradients approach their resolution limits, the GGR method
6 becomes inaccurate. Here, a minimum threshold for $\Delta[\text{CO}_2]$ of 2.4 ppm implies a maximum
7 wind speed of approximately 5 m s^{-1} .

9 3.2.3 Filter thresholds and data yield **Cuts in response to Ref. 1**

10 Of over 22,000 runs with measured $\Delta[\text{CO}_2]$, 9193 for the IFR site and 6912 for the U UW site
11 passed the $\Delta[\text{CO}_2]$ threshold value. The higher rejection rate for the U UW site is consistent
12 with CO_2 fluxes and gradients at that site being generally closer to zero than at the IFR site. A
13 few hundred runs were eliminated during grazing events. Further, ~~it cannot be assumed that~~
14 ~~the flux gradient approach holds in low turbulence conditions. Since~~ since the CO_2 fluxes for
15 runs with $\sigma_w < 0.112 \text{ m s}^{-1}$ were excluded and replaced with gap-filled values ~~anyway~~, such
16 runs were not used for the GGR method. Of the remaining runs, ~~46306 %~~ at the IFR and
17 ~~329613 %~~ at the U UW site, ~~those with~~ had to be discarded because of negative ~~K_{CO_2} were~~
18 ~~discarded, too,~~ K_{CO_2} , leaving 42433757 and 27232424 runs, respectively. The overall relative
19 data yield of the GGR method was thus 1917 % at the IFR site and 1211 % at the U UW site.

21 3.2.4 Footprint considerations

22 For the CO_2 fluxes measured by eddy covariance, the footprints were computed with the tool
23 of Neftel et al. (2008), which implements the model of Kormann and Meixner (2001). The
24 footprint contributions from the target surface were found $> 90 \%$ most of the time, at both
25 sites. ~~For details see Appendix B of~~ (Hunt et al. ~~(2015., 2016)~~).

26 Flux footprints depend on measurement height. For the trace gas fluxes using the GGR
27 method, the effective measurement height must be somewhere between the heights of the two
28 intakes (0.76 and 1.96 m). This intermediate height, regardless of how exactly it is specified,
29 is lower than the eddy-covariance measurement height (1.86 m), hence the footprint
30 contributions of the target surface to the CH_4 and N_2O fluxes are even larger than for the eddy

1 fluxes. Compared to the overall uncertainty of the fluxes, the footprint contributions from
2 areas outside the target surface are thus considered negligible, and no wind-direction filtering
3 was applied to the data.

5 3.2.5 Diurnal courses and daily means of fluxes

6 An example of CH₄ and N₂O fluxes obtained with the GGR method is shown in Fig. 5, along
7 with wind speed. Some negative flux values occur, and since negative diffusivities have been
8 explicitly excluded, these must be due to positive (upwards-increasing) gradients. Data yield
9 is higher at the IFR site, as ~~was~~ already noted to be the case throughout. At both sites the
10 range of N₂O fluxes is fairly consistent from one day to the next, the daily mean N₂O fluxes
11 are positive for all days shown, and the scatter is of the same magnitude as the mean. For the
12 CH₄ fluxes, the picture is less consistent: on some days (e.g. 14 Feb) the range is much larger
13 than on others (e.g. 17 Feb), the large range includes positive and negative fluxes, and on days
14 with such a large range the scatter is much larger than the mean and makes the latter's value
15 highly uncertain. The days with large scatter tend to be windier than those without, so in part
16 the large scatter may be due to relatively small CH₄ gradients, near the FTIR resolution limit,
17 being multiplied with large diffusivities, which in effect amplifies the random error of the
18 gradient measurements. This can of course occur for N₂O, too. Another possibility, more
19 specific to CH₄, is the existence of large sources, such as grazing animals, in nearby
20 paddocks. Temporal variations of wind direction may cause the emissions plumes from such
21 sources to contaminate the air sampled from either intake. Since the intakes are sampled
22 sequentially, the sign and magnitude of the contamination effect on the gradient would then
23 depend strongly on the exact timing of such a plume's passage. Lacking detailed information
24 of cattle presence in paddocks other than the target paddocks, we do not have any objective
25 criteria to decide which CH₄ flux records are contaminated. Hence, we only apply a crude
26 outlier filter, by removing fluxes that fall outside ± 5 times the SD of the whole dataset.

27 Assuming that no strong diurnal courses are expected for either CH₄ or N₂O fluxes, and that
28 the within-day variability of both is largely random, we assess seasonal variability from daily
29 means and their standard errors (SE). Days with fewer than four valid runs were excluded.
30 Daily fluxes were obtained on 6559 % and 4638 % of all days for the IFR and the UUW site,

1 respectively. These were spread reasonably evenly across all seasons (Figs. 6, 7), with the
2 lowest coverage in June (early winter).

3 At both sites, daily means of CH₄ fluxes using the GGR method were generally small
4 compared to their SE and thus often not significantly different from zero. No seasonal trends
5 were discernible, and no quasiperiodic patterns that could be associated with the rotational
6 grazing, either. The median daily CH₄ fluxes were 4.03.7 and 5.24.9 nmol m⁻² s⁻¹ at the IFR
7 and the UUW site, respectively, indicating small net emissions overall; the median SE of
8 these daily fluxes were 7.78.2 and 7.68 nmol m⁻² s⁻¹, respectively.

9 Daily means of N₂O fluxes from the GGR method also occurred with either sign (middle
10 panels of Figs. 6, 7); however, after 15 October 2013, when the modified switching cycle
11 (12 min per intake) was used, negative daily fluxes became rare. At the IFR site, daily N₂O
12 fluxes spanned the range ±5 nmol m⁻² s⁻¹, but the majority of values were of positive sign.
13 The median daily flux was 0.52 nmol m⁻² s⁻¹. This was greater than the median of the
14 associated SE values, of 0.3639 nmol m⁻² s⁻¹, and thus indicated significant net emissions
15 overall. At the UUW site, daily N₂O fluxes ranged from -1.2 to 31.9 nmol m⁻² s⁻¹. The
16 median flux was 0.2528 nmol m⁻² s⁻¹, and the median SE of the daily fluxes was
17 0.3529 nmol m⁻² s⁻¹, respectively. As with CH₄, it was difficult to detect any seasonal trends,
18 except for a slight tendency for larger variability throughout the irrigation seasons (which are
19 largely identical with the seasons of N inputs from grazing and fertiliser application). In
20 particular, there were no obvious responses of N₂O fluxes to soil moisture (bottom panels of
21 Figs. 6, 7).

22

23 **3.3 NSR method**

24 **3.3.1 NSR regressions**

25 For the NSR method, linear regression slopes for N₂O and CH₄ mole fractions versus CO₂
26 mole fraction were determined for each night in which runs with $\sigma_w < 0.1012$ m s⁻¹ occurred
27 (including only these runs in the regression). To be reliable, the linear relationship must have
28 a high R² and must be based on a sufficient number of runs, *n*. Therefore, minimum
29 thresholds for both parameters are required. During most low-turbulence nights, the
30 regressions of [N₂O] versus [CO₂] had R² values greater than 0.7, at both sites. There was no

1 relationship between R^2 and n , except that the scatter in R^2 seemed to increase with
2 decreasing n (not shown). For $[\text{CH}_4]$, the R^2 values of the NSR regressions versus $[\text{CO}_2]$
3 tended to be lower than for $[\text{N}_2\text{O}]$; their majority spread between 0.3 and 0.8, again
4 irrespective of n . Lacking any obvious cut-off values for R^2 and n , their choices must be made
5 pragmatically by balancing the reliability of the individual regression (in favour of high
6 thresholds) against the statistical power of including a large number of nights (in favour of
7 low thresholds). We used a minimum $R^2 = 0.4$ and a minimum $n = 4$, which leaves about
8 29 % of nights available for CH_4 and about 48 % for N_2O . **added in response to Ref. 1**

9 Fig. 8 shows the evolution of the NSR regression slopes over time. For three quarters of the
10 year, the N_2O vs CO_2 slopes show mainly short-term variations, but hardly any seasonal
11 trend; yet during the colder months May to July they increase to 3 to 5 times higher levels.
12 Both the short-term and the seasonal variations are very similar for the two sites. The CH_4 vs
13 CO_2 slopes are less well correlated between the two sites, and short-term variations dominate
14 throughout, while seasonal trends are absent.

15

16 3.3.2 Nocturnal CO_2 fluxes

17 The NSR method requires one representative CO_2 flux value for each night. This value was
18 obtained as the mean of the half-hourly fluxes from the gap-filled EC records. As a quality
19 check on the gap-filling, Fig. 9 compares the night-mean EC fluxes from the complete, gap-
20 filled record with night-means obtained from measured CO_2 fluxes using only runs with
21 $\sigma_w > 0.1012 \text{ m s}^{-1}$ (including only nights with at least 9 such runs available). At both sites, and
22 for all seasons, the two time series track each other well. The measured-only series appears at
23 times more scattered, but no systematic differences occur.

24 For the period from 23 September 2013 to 31 March 2014, mean nocturnal CO_2 fluxes at the
25 IFR site were also computed from the respiration chamber data. The ratio of the night-mean
26 EC fluxes to night-mean chamber fluxes was on average 0.87 (± 0.34 SD), and agreement was
27 generally best for the first few days following grazing events (matched by cutting of the grass
28 in the chambers).

29

3.3.3 Footprint considerations and nocturnal fluxes of CH₄ and N₂O

From footprint models it is known that the source area influencing a flux measurement in the atmospheric surface layer has a larger extent when stratification is stable than when it is unstable (Schmid, 1994; Kormann and Meixner, 2001). During nocturnal low-wind periods, stable stratification prevails, and so, comparatively large source areas can be expected for the NSR method. Further, the source area influencing a mole-fraction measurement extends farther than that influencing a flux measurement (Schmid, 1994). Since the NSR method combines measurements of a flux, that of CO₂, with measurements of the changes of a mole fraction, that of χ , over time, it can be hypothesised that its effective source area is somewhat larger than that of a direct flux measurement (e.g., by eddy covariance or the GGR method). For our setup geometry (Fig. 1), NSR measurements at the IFR site could in part be influenced by conditions over the dryland area, for wind directions either side of N, from 284° to 76°. Similarly, NSR measurements at the UUW site could in part be influenced from either of the two irrigated circles to the S and W, hence for any wind direction between 93° and 321°. Table 2 shows the differences in the median nocturnal fluxes between including and excluding periods with winds from the specified directional sectors, as well as the number of nights contributing to each median flux, N . The wind-direction filter is applied on a half-hourly basis, together with the low-turbulence filter ($\sigma_w < 0.1012 \text{ m s}^{-1}$), prior to the NSR regression and the filtering for $R^2 > 0.4$ and $n \geq 4$.

One effect of the exclusion of unsuitable wind directions is a considerable reduction of N , by about half. In addition (not shown), the number of runs contributing to each night, n , is also on average reduced. Of all nocturnal half-hours with low turbulence, 6663 % had unsuitable wind direction for the IFR site, and 7367 % for the UUW site. The reason for such high exclusion rates is a predominance of wind from the NW sector, due to katabatic flow from the Southern Alps. Unfortunately, those wind directions that were unsuitable at both sites (284° to 321°) were also the most common.

At the IFR site, the median CH₄ flux with wind-direction filtering is 63 % greater than without, ~~while~~ and the median N₂O flux ~~with filtering~~ is ~~unchanged~~ 5 % greater than without. At the UUW site, wind-direction filtering reduces the median CH₄ flux by 3538 % and the median N₂O flux by 2427 %. The effect of the filter at this site appears large enough to justify its application, despite the considerable reduction in data yield.

1 At both sites and for both gases, the means of the nocturnal fluxes were greater than their
2 medians, with SDs between 0.876 and 1.43 times the mean values. Due to these large relative
3 SDs, the means were quite sensitive to the presence (or exclusion) of outliers and are for that
4 reason not reported in Table 2.

6 3.3.4 Uncertainty of the CH₄ and N₂O fluxes

7 The uncertainty of the NSR regression slope is expressed as its standard error (SE). In relative
8 terms, this ranged from 2 % to 80 % of the regression slope, for both sites and gases. For CH₄,
9 the median relative errors of the slopes were ~~30 % at the IFR site and 29 % at the UUW~~
10 ~~siteboth sites~~. For N₂O, these median errors amounted to ~~21 %~~ 20 % at the IFR site and ~~17 %~~,
11 ~~respectively~~ % at the UUW site.

12 The uncertainty of the nocturnal CO₂ fluxes, from the gap-filled EC data, was specified as the
13 SEM for each night. This includes the contributions from random measurement error and
14 temporal variability during the night. Expressed as a relative error, it ranged from ~~10.4 %~~ to
15 ~~over 5033 %~~ of the flux but rarely exceeded ~~2015 %~~. The median relative errors were ~~54.2 %~~
16 and ~~5.74.3 %~~ for the IFR and the UUW site, respectively. The number of runs per night
17 contributing to the mean had a median of 21, at both sites. Multiplying the median relative
18 error of the night-mean flux with 21^{1/2}, the typical relative error of the individual half-hour
19 flux is obtained as ~~2419 %~~ and ~~2620 %~~ for the IFR and the UUW site. This appears realistic,
20 since nocturnal CO₂ flux errors in windy conditions are often estimated to be of order 20 %
21 (Moncrieff et al., 1996).

22 We thus estimate the SEM of the nocturnal CH₄ and N₂O fluxes by combining the SEM of the
23 nocturnal CO₂ fluxes with the SE of the regression slopes (using standard root-mean-square
24 propagation). The fluxes and their SEM are shown in Figs. 6 and 7, along with their
25 counterparts from the GGR method.

1 3.4 Combination of GGR and NSR methods

2 3.4.1 Comparison of the two methods

3 The GGR method provided a higher yield of daily flux values for CH₄ and N₂O than the NSR
4 method, but also the larger day-to-day variability (Figs. 6, 7). The N₂O fluxes from the NSR
5 method generally followed the trends of those from the GGR method well, at timescales of a
6 few days or longer. The same was true for the CH₄ fluxes at the U UW site (Fig. 7), with the
7 exception of two a couple of high outliers (in Nov 2012 and Jan 2013). For N₂O, the median
8 fluxes from the two methods were also in good agreement (for NSR 5.523 % and 134 %
9 greater than for GGR at the IFR and the U UW site, respectively).

10 About half of the CH₄ fluxes from the NSR method at the IFR site were considerably greater
11 than those from the GGR method, while the other half agreed well with the trend of the latter
12 (Fig. 6). The likely cause of the discrepancies for CH₄ is the presence of the cattle herd
13 elsewhere in the irrigated circle, acting as a strong source of CH₄ that was not uniformly
14 spread across the pasture surface in the same way as the sources of CO₂ and N₂O. During
15 stable surface-layer stratification, the CH₄ (and CO₂) emitted by the cattle would accumulate
16 in the surface layer and, with mean wind close to zero, slowly spread in all directions until,
17 eventually, being moved and dispersed by an intermittent flow event.

18 Without records of grazing events in paddocks other than those two containing our instrument
19 sites, there are no clear criteria to decide in which nights the NSR estimates of CH₄ flux are
20 significantly biased by cattle emissions and in which they are not. Histograms of the CH₄
21 fluxes for both sites (not shown) suggest that fluxes greater than 6080 nmol m⁻² s⁻¹ are very
22 likely outliers due to cattle emissions. We thus remove these from further analysis. This
23 reduces the number of nights, of 86101 and 7796 for IFR and U UW site (Table 2), to 7088
24 and 7595, respectively. However, the possibility remains that further nights are affected and
25 that the NSR estimates for CH₄ fluxes may be overestimated in these.

26

27 3.4.2 Construction of emissions budgets

28 The time series of daily CH₄ and N₂O fluxes can be used to construct emissions budgets for
29 longer periods, such as seasons or years. For this, we wish to make good use of the two
30 complementary methods. A simple approach is to take separate means for the GGR and the

1 | NSR method and then average ~~them~~these, giving equal weight to each method (we shall call
2 | this the “combined” approach). An alternative approach can be described as follows: first,
3 | average GGR and NSR means on a daily basis for each 24-h period in which both estimates
4 | are available; then, for 24-h periods in which only one method produced a valid mean flux,
5 | use that flux; finally, take the overall mean of this merged time series (we shall call this the
6 | “merged” approach). The merged approach gives more weight to the method with the greater
7 | data yield.

8 | We explore these two approaches for the first year of the dataset (17 August 2012 to
9 | 16 August 2013). With both approaches, we estimate the uncertainty by propagating the SE of
10 | all day-means/night-means into an SE value for the annual mean flux. The results are
11 | summarised in Table 3. All flux values in the table are positive, indicating net annual
12 | emissions of the two trace gases. Data yields (N) are included in the table; for the combined
13 | approach no N column is given because the individual N for the GGR and NSR method apply.

14 | For CH₄ at the IFR site, the NSR method estimates 5 times greater emissions than the GGR
15 | method, despite the outlier-filtering described above. Since the data yield of the GGR method
16 | was 43 times greater, the relative weighting of the two methods matters; consequently, the
17 | combined approach results in an emissions estimate that is ~~about twice~~1.6 times that of the
18 | merged approach. By contrast, for CH₄ at the UUW site, GGR and NSR estimates differ ~~only~~
19 | ~~by about 20 % and their propagated SE ranges overlap~~by less than a factor 2; consequently,
20 | the combined and merged approach agree ~~well with each other~~within 10 %.

21 | For N₂O at the IFR site, the annual NSR mean exceeds the annual GGR mean by a factor ~~of~~
22 | ~~about two~~2.6. This is a considerably greater difference than was found for the medians (of
23 | this year as well as of the whole dataset). The GGR mean includes ~~about 2/362 %~~
24 | of the year, while the NSR mean is based on ~~about 1/338 %~~
25 | of all nights. Both datasets are evenly spread across the seasons (Fig. 6). The GGR dataset contains a surprisingly large
26 | number of negative daily values in Dec 2012 and Jan 2013 while the NSR dataset does not
27 | contain negative values, and the different annual means for the two methods are largely
28 | caused by this. The combined approach results in an annual flux estimate that is ~~2923~~2923 %
29 | greater than the estimate from the merged approach, in which the NSR data have less weight.

30 | For N₂O at the UUW site, the annual NSR mean exceeds the annual GGR mean by 5457 %.
31 | Again, this is a greater difference than that for the medians. The data yields for both methods
32 | are smaller than at the IFR site, but roughly ~~in about~~ the same proportion with each other.

1 | ~~Consequently, the~~The combined approach results in a ~~145~~ % greater annual flux estimate than
2 | the merged approach, and the ranges indicated by the propagated daily SE are marginally ~~non-~~
3 | overlapping. Again, the GGR dataset contains some negative daily means (while the NSR
4 | dataset does not); these occurred mostly in Oct-Nov 2012, not at the same time as for the IFR
5 | site.

6 7 8 | **4 Discussion**

9 | **4.1 Performance of the GGR method**

10 | The GGR and NSR method exploit complementary datasets: for the former, only runs are
11 | used in which a set turbulence threshold is exceeded, for the latter only runs in which the
12 | opposite is true. The data yield of each method is thus dependent on the general wind climate
13 | of the site. However, each method also requires careful screening with other suitability
14 | criteria. For the GGR method, we have demonstrated that the main one is to ensure that the
15 | mole-fraction gradients of the reference gas (here CO₂) are a few times larger than instrument
16 | resolution, in order to contain random scatter within tractable limits. An implication of this
17 | requirement is that very high wind speeds tend to be less suitable than moderate wind speeds,
18 | because the former dilute emitted gases more quickly and thus reduce their gradients. The
19 | exact choice of filtering criteria must balance reliability (constraining random error) with
20 | availability (obtaining enough data coverage).

21 | Even with appropriate filtering, run-to-run variability of the GGR method is large, because
22 | the random errors of three measured variables (two gas mole-fraction gradients and the
23 | reference gas flux) combine, and partly because the two gas mole fractions are measured
24 | sequentially, not simultaneously (although usage of ballast volumes reduces the effects of
25 | timing mismatch somewhat). The random error of a daily mean flux depends, then, on the
26 | number of acceptable runs, and may thus be large on some days and small on others. In our
27 | experiment, we obtained acceptable daily mean fluxes for approximately ~~half~~40 % of all days
28 | at one site and ~~two-thirds~~60 % at the other. This was sufficient for adequate coverage of all
29 | seasons and the estimation of annual budgets of gas emissions.

1 The large uncertainty of individual runs found with the GGR method is similar to that found
2 with the momentum-based aerodynamic method (Phillips et al., 2007). The data yield of the
3 former is probably smaller than that of the latter, because suitably-large [CO₂] gradients
4 (required for GGR) may occur somewhat less often than reliable measurements of friction
5 velocity (required for the aerodynamic method). However, the GGR method avoids any
6 assumptions on Schmidt number and stability dependence and its diffusivity estimates are
7 therefore more defensible for the purpose of computing trace gas fluxes.

8 For both trace gases, negative flux values (uptake) were occasionally observed with the GGR
9 method. In the case of CH₄, the negative fluxes were spread across all seasons, at both sites.
10 They are thus considered as manifestations of the large random variability. By contrast,
11 negative fluxes of N₂O were occasionally clustered in certain periods. These cluster periods
12 were not synchronous at the two sites. During these, the mole-fraction gradients with reversed
13 sign occurred consistently over many successive cycles, suggesting that they had a true cause,
14 rather than occurring randomly. However, the NSR method did not indicate N₂O uptake
15 during these periods. While we cannot rule out the possibility that the negative N₂O fluxes
16 from the GGR method were artefacts, we have no indication from the data themselves, or our
17 records of instrument operation and farm management, why they might have been.

18

19 **4.2 Performance of the NSR method**

20 The NSR method relies on the occurrence of nocturnal calm periods of a few hours duration.
21 Such periods do not occur every night. When they do, the method is technically robust, since
22 the gas mole-fraction changes over time tend to be large and can be well-resolved. A crucial
23 assumption of the NSR method is the co-location, in space and time, of the sources or sinks of
24 the trace gas of interest and the reference gas. In our experiment, the results for N₂O were
25 very consistent with those from the GGR method, so we conclude that for N₂O and CO₂ over
26 pasture this assumption holds well enough, as it did in the pioneering experiment of Kelliher
27 et al. (2002). The results for CH₄ were less consistent, and we attribute that to the presence of
28 cow herds in the larger surroundings of the measurement site. These constituted strong
29 additional sources, of both CH₄ and CO₂ but with a very different emissions ratio to that valid
30 for the pasture, and therefore occasionally (depending on intermittent winds) impacting on the
31 correct retrieval of the latter.

1 The footprint of the NSR method has a rather large spatial extent, compared to the GGR
2 method. Because of the requirement of co-location of sources/sinks, one needs to exclude
3 periods when the extension of the target surface in the wind direction is too short. In our
4 experiment, the prevalence of katabatic flow from a certain directional sector led to the
5 exclusion of 5040 % or more of otherwise suitable calm nights. Such high exclusion rates
6 could be avoided by maximising the fetch in the main regional upslope direction (but in our
7 case, this would have conflicted with the requirements of the dual-site setup and was thus
8 impossible). Here, the NSR method provided fewer night-mean flux estimates than the
9 number of daily means obtained with the GGR method; however, this does not need to be the
10 case in general, since the data yield of both methods depends on local wind climate and
11 footprint considerations.

12 The NSR method requires reliable flux estimates for the reference gas. We have here explored
13 a novel approach to this end, by using gap-filled long-term time series of CO₂ flux records,
14 instead of chamber measurements. Hence, the actual flux values used for the NSR-suitable
15 nighttime periods were modelled, but strongly based on measured fluxes in the surrounding
16 windier periods. They were validated against chamber measurements and found to provide
17 adequate estimates of the nocturnal mean respiration flux, with a relative uncertainty that was
18 usually smaller than that of the NSR regression slope and therefore did not impact negatively
19 on the success of the NSR method.

20

21 **4.3 Annual means of CH₄ and N₂O emission rates**

22 Either of the two methods, GGR and NSR, can provide annual budget estimates of trace gas
23 fluxes on its own, provided that these fluxes do not undergo strong systematic diurnal
24 variations. If they did, then biases would be likely, since the GGR method yields more data
25 during the day than at night and the NSR method is explicitly nocturnal only. We thus
26 strongly recommend to combine the two methods. As they are applicable in mutually-
27 exclusive conditions (well-developed turbulence and calm, respectively), their combination
28 optimises data usage.

29 We tested two approaches to combine the GGR and NSR results: either combining their
30 separate annual means, or merging the two time series into one before computing a joint
31 annual mean. Ideally, the two approaches should not differ by much. Here, we found that to

1 be the case for the CH₄ fluxes at the U UW site. However, when the results from the GGR and
2 NSR methods show systematic differences, then some operator judgment is required whether
3 that reflects true day-night differences of the fluxes or whether one of the two methods is
4 likely to be biased. In the former case, combining GGR and NSR means would be adequate,
5 as that would give similar weights to daytime and nighttime data. In the latter case, the likely
6 cause of bias needs to be assessed and the decision how to weigh the GGR and NSR results
7 must be based on that assessment.

8 For the CH₄ results at the IFR site, we suspect that the mean fluxes from the NSR method are
9 overestimated in many nights, due to the presence of cows emitting CH₄ elsewhere on the
10 irrigated-pasture circles. Thus, for CH₄ we decide to give high weight to the GGR method, by
11 using the results from the merged approach: ~~7.0 (±0.89) and. This yields identical emission~~
12 ~~rates of 8.29 (±0.9179) nmol m⁻² s⁻¹ for the IFR and the U UW site, respectively two sites~~
13 (Table 3). ~~With this approach, the mean annual emission rate from the IFR site is slightly, but~~
14 ~~not significantly, smaller than that from the U UW site.~~ If we used the combined approach
15 instead, the mean emission rate from the IFR site would appear as the significantly greater
16 one.

17 For annual N₂O fluxes, the mean from the NSR method was somewhat greater than that from
18 the ~~GSR~~GGR method, at both sites. We found the differences to originate mainly from certain
19 periods in which the GGR method repeatedly indicated negative fluxes, while the NSR
20 method did not. It is possible that during these periods the pasture acted as an N₂O sink during
21 daytime and as a source during the night; Hörtnagl and Wohlfahrt (2014) observed such
22 behaviour in the spring seasons of two consecutive years and in autumn for one of these
23 years. Therefore, we decide to use the combined approach for N₂O, giving equal weight to the
24 GGR and NSR method despite the greater data yield of the former. Considering high N₂O
25 fluxes as undesirable, this decision errs on the side of caution and produces somewhat greater
26 emission rates than would be obtained with the merged approach. These emission rates are
27 ~~0.5758 (±0.019020)~~ and ~~0.4038 (±0.018)~~ nmol m⁻² s⁻¹ for the IFR and the U UW site,
28 respectively (Table 3). Hence, the emission rates from the IFR site were significantly greater
29 than those from the U UW site, by ~~4253~~ %. This is in line with expectations, given the greater
30 N inputs from fertiliser application and excreta deposition at the IFR site.

31

1 4.4 CH₄ fluxes from managed grasslands

2 Agricultural soils, when not waterlogged, are expected to be weak sinks for CH₄ (Smith et
3 al., 2003), and chamber studies have tended to confirm this expectation. For example, Imer et
4 al. (2013) reported net uptake rates for three managed ungrazed grassland sites in
5 Switzerland; Savage et al. (2014) found annual CH₄ uptake of 0.2 g C m⁻² for an alfalfa field
6 in North Dakota, using year-round auto-chamber sampling. In New Zealand, Li and Kelliher
7 (2007) reported net uptake rates of 0.14 and 0.05 g C m⁻² yr⁻¹ for a well-drained and a poorly-
8 drained dairy-pasture soil, situated 300 m apart. By contrast, the results reported here indicate
9 both the IFR and the UUW site to be CH₄ sources, with similar annual net emissions (for the
10 year starting 17 August 2012) of 2.6 and 3.14 g C m⁻², respectively. Even if one discarded
11 the NSR results completely on suspicion of CH₄ contamination by animal herds in the wider
12 surroundings, the GGR datasets alone still have positive means and medians, at both sites.
13 Since the daytime footprint extensions for the GGR method were of order 300 m or less, we
14 can be confident that the GGR results were generally representative for pasture surfaces free
15 of grazing animals.

16 Other micrometeorological studies of grasslands are useful to compare with our CH₄ results
17 only where grazing events were excluded from the data, where the soils were not peaty and
18 where the footprint contained no open water surfaces. Using eddy covariance, Hörtnagl and
19 Wohlfahrt (2014) found, at an alpine meadow that was cut three times per year, CH₄ emission
20 rates of order 1 nmol m⁻² s⁻¹; these were very consistent throughout all snow-free seasons.
21 For a grassland site after restoration, Merbold et al. (2014) report annual CH₄ emissions of
22 3.53 g C m⁻², 14 and 35 % greater than at very similar to our IFR and UUW site,
23 respectively two sites, and daily mean emissions were in the majority of positive sign in every
24 month. Griffith et al. (2002), using the flux-gradient method over lucerne pasture in New
25 South Wales, reported mean and median daytime fluxes of 2.95 and 2.85 nmol m⁻² s⁻¹, and
26 even though these were not significantly different from zero due to the short duration of that
27 experiment (3 weeks), the sign and magnitude agree with those from the other
28 micrometeorological studies. It appears that net CH₄ emissions from managed grasslands are
29 not uncommon, which is at odds with those chamber experiments that demonstrated CH₄
30 oxidation. A possible explanation is that frequent N fertilisation inhibits the oxidation process
31 and/or stimulates CH₄-producing microbes in the soil (Price et al., 2004). Li and Kelliher
32 (2007) found that urine application reduced net CH₄ uptake significantly, both for a well-

1 drained and a poorly-drained soil. We note, though, that the net uptake rates observed for each
2 of their treatments were at least one magnitude smaller than the mean emission rates at our
3 sites. The same is true for an experiment with intact soil cores from European sites (Schaufler
4 et al., 2010), where the cropland and grassland soils showed CH₄ fluxes smaller than
5 0.06 nmol m⁻² s⁻¹, some with net uptake and some with net emissions, and without
6 dependence on soil moisture or temperature. Another possible factor favouring CH₄ emissions
7 may be the soil compaction caused by intensive grazing events, provided that leads to an
8 increased occurrence of anaerobic microsites in the soil. Whatever the exact mechanisms, the
9 field-scale CH₄ flux measurements here and elsewhere suggest that on managed pastures, CH₄
10 oxidation rates in the soil can be overwhelmed by concurrent emission processes.

11

12 **4.5 N₂O emissions and nitrogen inputs**

13 The time series of N₂O fluxes in Figs. 6 and 7 are, overall, relatively steady. In Fig. 6, there is
14 no quasiperiodic pattern that would indicate a response to events of strong N input (grazing or
15 fertilisation). There is also an almost complete lack of strong episodic events. Such events
16 have often been reported to closely follow rainfall or irrigation events (e.g. van der Weerden
17 et al., 2013), and they are found to occur in a relatively narrow range of soil moisture that is
18 site-specific. Balaine et al. (2013) present evidence that the driving variable for peak N₂O
19 emissions is the relative soil gas diffusivity. Here, at the IFR site throughout the milking
20 season, it appears that VWC was generally too low to reach the conditions required for peak
21 emissions. The highest VWC occurred over winter 2013 (June to August), generally between
22 0.4 and 0.55 m³ m⁻³ (equivalent to 60 and 83 % water-filled pore space, respectively, given a
23 porosity of 66 % in the soil layer from 0 to 10 cm depth). During this winter period, the N₂O
24 fluxes appeared particularly small and steady, at 0 to 0.5 nmol m⁻² s⁻¹. This was also a period
25 in which no fertilisation or grazing events occurred. The relative steadiness of N₂O emission
26 rates throughout all seasons is probably largely due to the well-drained soil. This feature
27 distinguishes this intensively managed pasture site from others (Phillips et al., 2007) where
28 emissions were of a more episodic nature and more obviously responding to irrigation events.

29 In Table 4, we relate the N₂O emissions at the IFR site to the N inputs, for the same year as in
30 Table 3. The N inputs are obtained as follows. Fertilisers were applied by a certified
31 commercial contractor who ~~also~~ recorded the amounts per area; we assume a relative error of

1 7 % from expert judgment. The N inputs from cattle excreta can be accounted for as the N
2 intake by the animals reduced by the N retention in liveweight and the N ~~contents~~content of
3 milk produced, under the assumption that all excreted N is returned to the irrigated-pasture
4 circle either in-situ as dung and urine or by effluent application. ~~Following~~ Hunt et al. ~~(2015)~~
5 ~~calculated~~(2016), the dry-matter intake of the cattle herd for the 2012/13 milking season
6 ~~as was~~ 1.232167 (± 0.034026) kg m⁻². ~~(90 % grazed pasture, 10 % barley supplements)~~. With
7 an N ~~contents~~content of 2.7 (± 0.2) % for ryegrass/clover pasture (Mills and Moot, 2010), the
8 N intake ~~was 33.3~~amounted to 31.5 (± 2.64) g N m⁻². The total liveweight gain of the herd
9 over the milking season was estimated as 52,200 kg ~~(Hunt et al. 2015)~~. ~~With a pasture, based~~
10 ~~on national statistics (DairyNZ, 2013) and the age structure of the herd. Dividing by the total~~
11 ~~grazed~~ area of ~~263~~the farm (328 ha), and assuming an N ~~contents~~content of 3.6 % for meat
12 (AMC, 2014), the N retention in liveweight per pasture area was 0.757 g N m⁻² (~~21.8~~ % of N
13 intake). The milk production of the farm for 2012/13 was 3.96 * 10⁶ L. Assuming that the
14 milk contained 5.8 (± 0.5) g N L⁻¹ (Woodward et al., 2011), the amount of N exported in milk
15 was 97.0 (± 0.76) g N m⁻² (~~2722~~ % of N intake). The amount of N returned in excreta thus
16 results as 23.69 (± 2.75) g N m⁻².

17 The figure for the N₂O emissions in Table 4 comes from the combined approach (see Section
18 4.3), converted from molar units (0.5758 nmol m⁻² s⁻¹) to mass units and integrated over a
19 year, yielding 0.5051 g N m⁻². The uncertainty of this value is assumed as 10 % in relative
20 terms. This is greater (about 3 times) than the propagated SE in Table 3, partly to account for
21 the mismatch between approaches as discussed above, and partly to allow for the possibility
22 that the spatial variability in the whole irrigated circle may have been larger than in the
23 footprint of our measurement site. Dividing the N₂O emissions by the N inputs, the emission
24 factor results as 1.4921 (± 0.15) %. We consider this as an upper estimate because some
25 fraction of the measured emissions may be natural background emissions. In a chamber
26 experiment in February/March 2014, N₂O fluxes from the control plots (which were irrigated
27 but received no N) were consistently about 0.5 mg N m⁻² d⁻¹, over 35 d, (Jennifer Owens,
28 pers. comm.). Integrated over a year, this would amount to 0.18 g N m⁻². If we tentatively use
29 this value as “background” emissions (even though a fraction of it may still have occurred in
30 response to preceding fertilisations), then the net N₂O emissions above background would
31 have been 0.3233 g N m⁻² and the emission factor 0.7678 %. This value is very close to the
32 mean emission factor for dairy cattle excreta, of 0.73 %, from a statistical analysis of 125
33 field trials in New Zealand (Kelliher et al., 2014). With or without the background included,

1 the observed emission factor is also compatible with the value of 1 % recommended for
2 national greenhouse-gas inventories (IPCC, 2006).

3 At the U UW site, there was no fertiliser applied during the year starting 17 Aug 2012. There
4 was one winter grazing by 200 cattle in May; Fig. 7 shows a few elevated points in the time
5 series of the N₂O flux that may have occurred in response to that. There had also been a
6 winter grazing in June 2012, before our recording began. Some of the observed emissions
7 early in the budget year would have originated from the N deposited during that event,
8 possibly amplified by the effects of trampling (Pal et al., 2014). It is therefore likely that the
9 annual emissions at the U UW site, of 0.3534 g N m⁻², were also greater than natural
10 background emissions.

11 The annual N₂O emissions at our two sites are comparable to those from grassland sites
12 elsewhere in the world. Phillips et al. (2007) measured N₂O fluxes from flood-irrigated,
13 rotationally-grazed pasture in SE Australia. For two consecutive years, they obtained annual
14 emissions of 0.55 and 0.44 g N m⁻², respectively, bracketing the result for the IFR site.
15 Burchill et al. (2014) found, for a rotationally-grazed and intensively fertilised ryegrass
16 pasture in Ireland, annual N₂O emissions over 4 years to range from 0.44 to 3.44 g N m⁻². The
17 annual emissions from the IFR site are within this range, near the lower end. Burchill et al.
18 (2014) also measured the annual emissions from ungrazed, unfertilised ryegrass pasture.
19 These ranged from 0.17 to 0.63 g N m⁻², i.e. from about half to about twice the annual
20 emissions from the U UW site. Soussana et al. (2007) reported annual N₂O emissions from
21 10 European grassland sites to range from -0.025 to 0.687 g N m⁻²; management practices
22 and fertiliser application rates at these sites varied considerably. Imer et al. (2013) found
23 annual emissions for three managed ungrazed grassland sites from 0.13 to 1.13 g N m⁻².

24

25 **4.6 Comparison to other methods**

26 Traditionally, the most common methods to measure trace gas fluxes from or to soils and
27 vegetation have been chamber methods. Their main advantages are high accuracy of the
28 individual sampling, and often low costs (Denmead, 2008). One of their main drawbacks is
29 limited coverage in time (due to operational constraints), though modern autochamber
30 systems with robust field-worthy gas analysers are largely overcoming this disadvantage (van
31 der Weerden et al., 2013; Savage et al., 2014). The other main drawback is that they do not

1 operate at the field scale, which is of particular relevance where gas emissions occur highly
2 concentrated from preferential spots, such as patches of animal excreta. Micrometeorological
3 methods, by contrast, integrate over this spatial variability, and they are in principle
4 continuous in time, except for certain filtering requirements, such as for unsuitable wind
5 direction or insufficient turbulence.

6 For CH₄ and N₂O, the most commonly used micrometeorological method nowadays is eddy
7 covariance, thanks to the advent of a number of fast and precise gas analysers based on laser
8 technologies. Drawbacks of EC are that these analysers are expensive and must be dedicated
9 to a single site, that there are often issues with instrument noise and signal resolution,
10 particularly when fluxes are small for extended periods, and that for periods of small fluxes,
11 the accuracy depends strongly on corrections for density effects and cross-sensitivities (Neftel
12 et al., 2010). Since EC relies on stationary turbulent flow in the surface layer, data from calm
13 nights must be discarded. In this respect, EC is very similar to the GGR method and other
14 variants of flux-gradient methods. Our approach to combine precise mole-fraction
15 measurements of trace gases of interest with EC measurements of CO₂ offers the following
16 advantages over direct EC measurements of the trace gases. First, it allows to measure at
17 more than one site with the same gas analyser (provided the sites are not too far apart).
18 Second, it allows to use a multi-gas analyser, such as the FTIR; in fact this is the ideal choice
19 because the mole fractions of the gas of interest and of CO₂ are measured in the same air
20 sample. Third, by then using GGR and NSR as complementary methods to compute the trace
21 gas fluxes, the sampling includes periods of well-developed and low turbulence and leads to a
22 higher data yield than EC or GGR alone. Of course, the NSR method could potentially also be
23 applied as a complement to direct EC measurements, provided that the EC system includes
24 CO₂ flux measurements (which is routinely the case) and that these are taken close enough to
25 the ground to keep the footprint of the NSR method representative for the surface of interest
26 (which may be less common).

27 The GGR and NSR method each require some optimisation of data filtering criteria: the GGR
28 method for sufficient resolution of CO₂ mole-fraction gradients and positive gas diffusivities,
29 and the NSR method for reliability of nocturnal linear regressions and possibly for suitable
30 wind direction (site-dependent). These criteria appear relatively straightforward, compared to
31 the suite of data quality filters required for the eddy-covariance method. Further, the GGR and
32 NSR method each employ simple algebraic relationships for gas ratios, which do not require

1 further corrections; in this, they are easier to implement than the eddy-covariance method
2 with its rather complex correction procedures involved in the computation of minor trace gas
3 fluxes.

4

5

6 **5 Conclusions**

7 Continuous year-round measurements of the fluxes of CH₄ and N₂O at two neighbouring,
8 contrasting pasture sites were obtained with the combination of GGR and NSR methods, both
9 using CO₂ as the reference gas (tracer). The CH₄ and N₂O fluxes resembled those from other
10 managed grasslands measured with the eddy-covariance or the flux-gradient method. The
11 combination of GGR and NSR methods can thus serve as a viable alternative to eddy
12 covariance and is particularly attractive in paired-site setups. However, the NSR method
13 should be applied with caution for trace gases that have strong sources or sinks not co-located
14 with those for the reference gas. Land-use patterns in the surrounding area, as well as regional
15 topography and climate, therefore influence the yield of usable data.

16 A novelty introduced here, different to the original concept of the NSR method (Kelliher et
17 al., 2002), is the usage of gap-filled CO₂ flux time series from eddy covariance, instead of
18 CO₂ fluxes from respiration chambers. Mean nocturnal CO₂ fluxes from this approach agreed
19 well with those from chambers, proving its validity.

20 For N₂O, the fluxes obtained with the GGR and NSR method were in reasonable agreement
21 with each other, and they were also fully compatible with the results from a wealth of
22 chamber studies and with recommended emission factors for N₂O emissions from dairy
23 pasture. The combination of GGR and NSR is thus a reliable option to obtain long-term
24 budgets of the N₂O exchange of ecosystems on flat land, with the advantage of effective
25 spatial integration of the source area (which is not guaranteed for chamber systems).

26 For CH₄, both the GGR and NSR method indicated net emissions from both pasture sites.
27 These were similar to emissions at other grassland sites measured with micrometeorological
28 methods. However, chamber measurements on grassland often show CH₄ fluxes that are one
29 or two magnitudes smaller and in the opposite direction, indicating CH₄ oxidation in the soil.
30 It is at this stage unclear whether these discrepancies have their origin in the different
31 methods (e.g. the different spatial coverage with chambers and micrometeorological methods)

1 or in different management practices (e.g. fertilisation amounts and frequency, disturbance
2 from animal treading).

3

4

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6

1 **Tables**

2

3 **Table 1.** Results of repeated mole-fraction measurements, with the FTIR spectrometer, of air
 4 from a gas cylinder with near-ambient composition (quality control). The two switching
 5 cycles are described in Section 2.2.1. Numbers in parentheses are standard deviations, from n
 6 successive cell fills.

7

Switching cycle	n	[N ₂ O] (ppb)	[CH ₄] (ppb)	[CO ₂] (ppm)
Original – 6 min	38	384.07 (0.28)	1805.22 (0.58)	389.53 (0.15)
Modified – 12 min	43	384.53 (0.14)	1808.04 (0.20)	389.79 (0.12)

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11 **Table 2.** Medians of nocturnal fluxes computed with the NSR method, as well the number of
 12 nights (N) contributing to the median, for two wind-direction selections: all included or
 13 unsuitable ones excluded. Directions are considered unsuitable for the IFR site when the
 14 dryland area is upwind, and unsuitable for the UUW site when either of two neighbouring
 15 irrigated circles is upwind. See text for specification of the directional sectors.

16

Trace gas	Site	all wind directions		unsuitable directions excluded	
		median flux (nmol m ⁻² s ⁻¹)	N	median flux (nmol m ⁻² s ⁻¹)	N
CH ₄	IFR	<u>23.425.6</u>	<u>159172</u>	<u>24.826.3</u>	<u>-86101</u>
CH ₄	UUW	<u>41.914.0</u>	<u>132164</u>	<u>7.758.73</u>	<u>7796</u>
N ₂ O	IFR	<u>0.553604</u>	<u>259293</u>	<u>0.554637</u>	<u>132165</u>
N ₂ O	UUW	<u>0.371396</u>	<u>240279</u>	<u>0.285289</u>	<u>-99131</u>

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2 **Table 3.** Estimates of mean annual fluxes, for the period 17 Aug 2012 to 16 Aug 2013, from four approaches: GGR method, NSR method,
 3 mean of the annual means of GGR and NSR, and annual mean of daily values obtained from merging the two methods. Numbers in
 4 parentheses are error estimates from propagation of daily standard errors. *N* is the number of nights contributing.

5

Trace gas	Site	GGR method		NSR method		GGR and NSR means combined	GGR and NSR merged daily	
		mean flux (nmol m ⁻² s ⁻¹)	<i>N</i>	mean flux (nmol m ⁻² s ⁻¹)	<i>N</i>	mean flux (nmol m ⁻² s ⁻¹)	mean flux (nmol m ⁻² s ⁻¹)	<i>N</i>
CH ₄	IFR	<u>4.37</u>	<u>258</u>	<u>22.7</u> <u>24.3</u>	<u>627</u>	<u>1314.5</u> (0.8 <u>176</u>)	<u>7.8.9</u> (0	<u>2782</u>
		(0. <u>9992</u>)	<u>224</u>	(1. <u>2822</u>)	<u>7</u>		(<u>0.89.79</u>)	<u>59</u>
CH ₄	U UW	<u>6.7.8</u>	<u>178</u>	<u>-12.9.5</u>	<u>668</u>	<u>9.8.7</u> (0. <u>6365</u>)	<u>8.29</u> (0. <u>9479</u>)	<u>2182</u>
		(1. <u>4405</u>)	<u>147</u>	(0. <u>6077</u>)	<u>4</u>			<u>01</u>
N ₂ O	IFR	<u>0.3632</u>	<u>258</u>	<u>0.7783</u>	<u>412</u>	<u>0.5758</u> (0. <u>019020</u>)	<u>0.4447</u>	<u>2862</u>
		(0. <u>030034</u>)	<u>225</u>	(0. <u>022021</u>)	<u>137</u>		(0. <u>026024</u>)	<u>73</u>

N ₂ O	UUW	<u>0.3230</u>	<u>178</u>	<u>0.4947</u>	<u>801</u>	<u>0.4038</u> (0.018)	<u>0.3536</u>	<u>2232</u>
		(<u>0.030032</u>)	<u>147</u>	(<u>0.020018</u>)	<u>08</u>		(<u>0.024022</u>)	<u>11</u>

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Table 4. Nitrogen budget terms for the IFR site for the year 17 Aug 2012 to 16 Aug 2013. The N₂O emissions value is from the combined GGR and NSR methods, but with a larger uncertainty estimate to account for method discrepancy and potential spatial variability across the irrigated circle. The resulting emission factor is considered a “maximum” because the measured emissions include naturally-occurring background emissions.

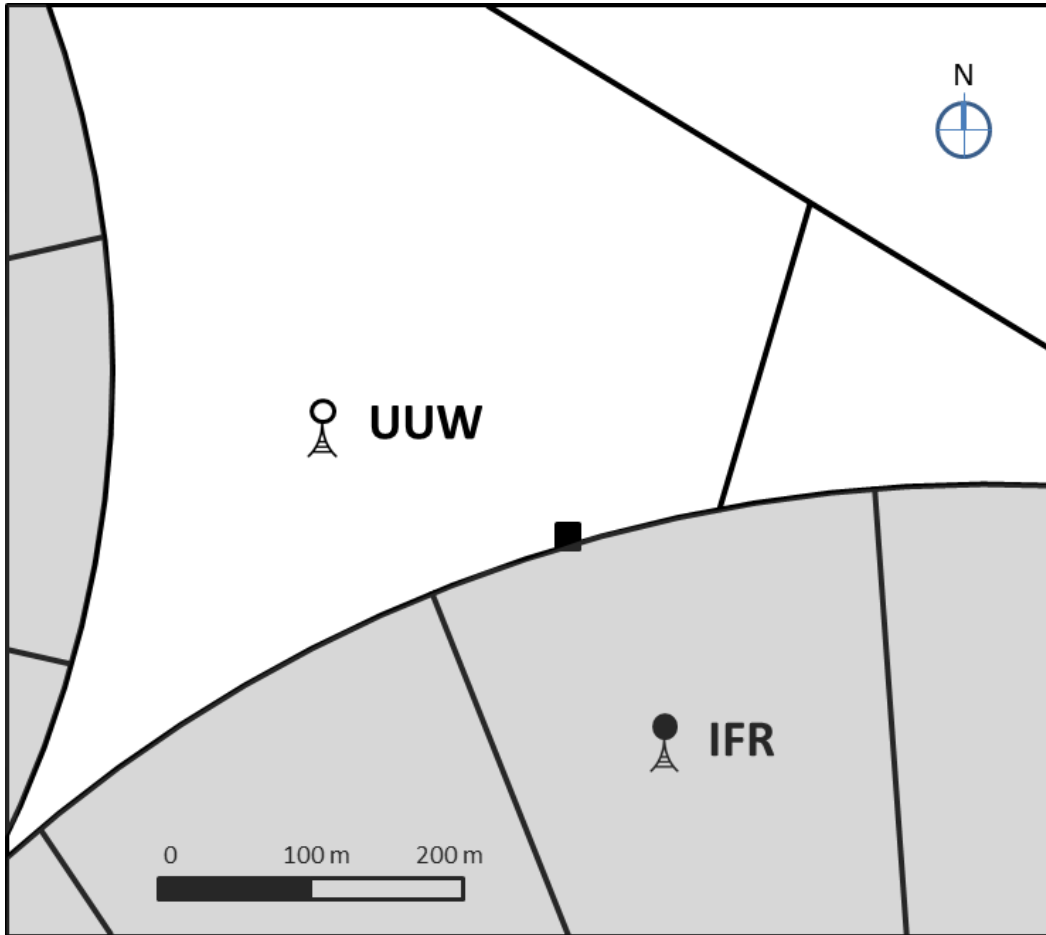
Annual N inputs (g N m ⁻²)	
Urea applied	18.3 (1.3)
Excreta (dung, urine, effluent)	23.69 (2.75)
Total	41.9 (3.042.2 (2.8))
Annual N ₂ O emissions (g N m ⁻²)	0.5051 (0.05)
Maximum emission factor for N ₂ O (%)	1.4921 (0.15)

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1 **Figures**

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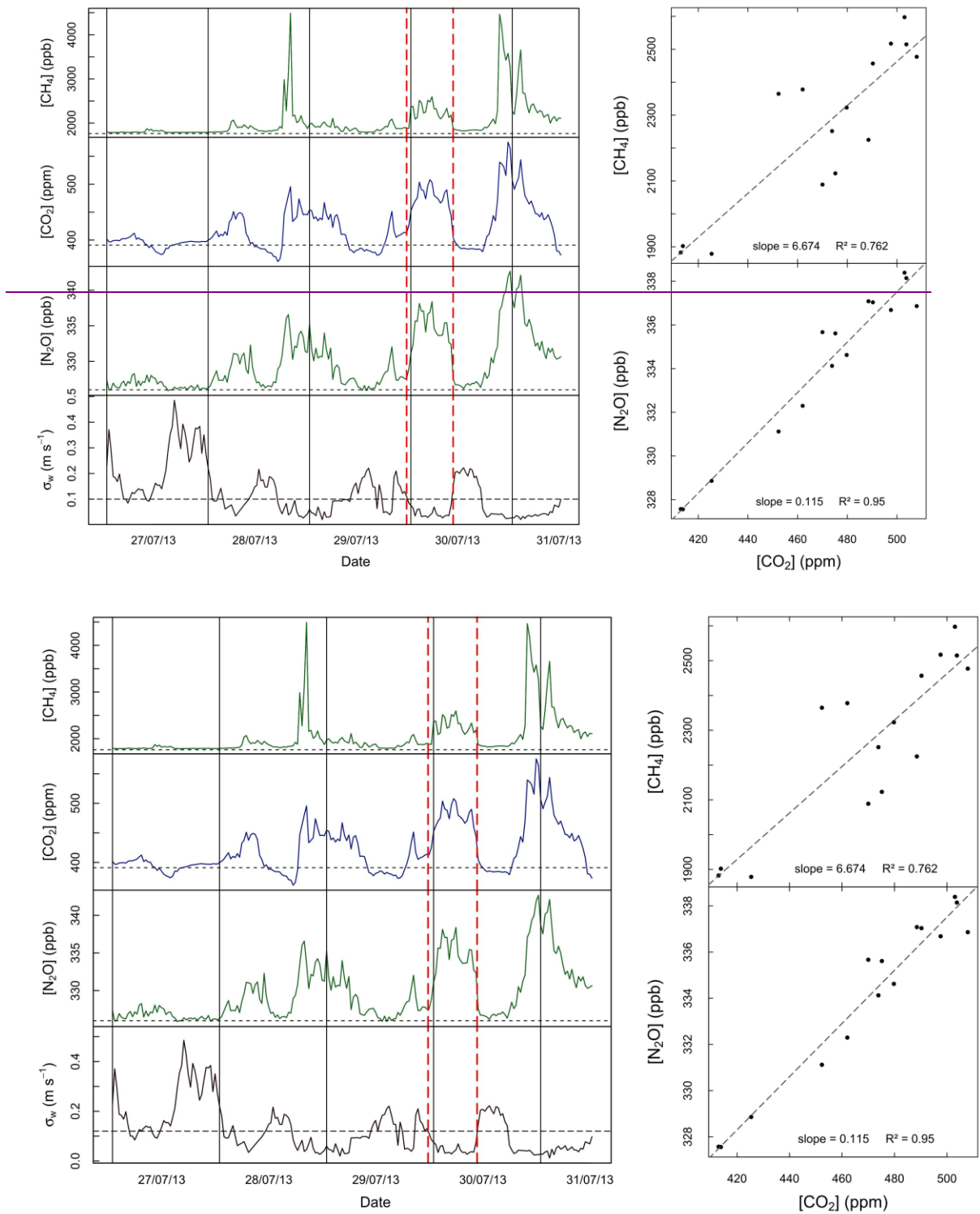
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5 **Figure 1.** Schematic of the experimental area. The shaded areas represent parts of pivot-
6 irrigated intensively-managed circles of dairy pasture. The white areas are not irrigated. At
7 each of the labelled locations, UUW (unirrigated, unfertilised, winter-grazed) and IFR
8 (irrigated, fertilised, rotationally-grazed), CO₂ fluxes were measured by eddy covariance, and
9 air was sampled from two heights for multi-gas mole-fraction measurements with an FTIR
10 spectrometer. The spectrometer was located in a temperature-controlled hut, indicated with a
11 black square.

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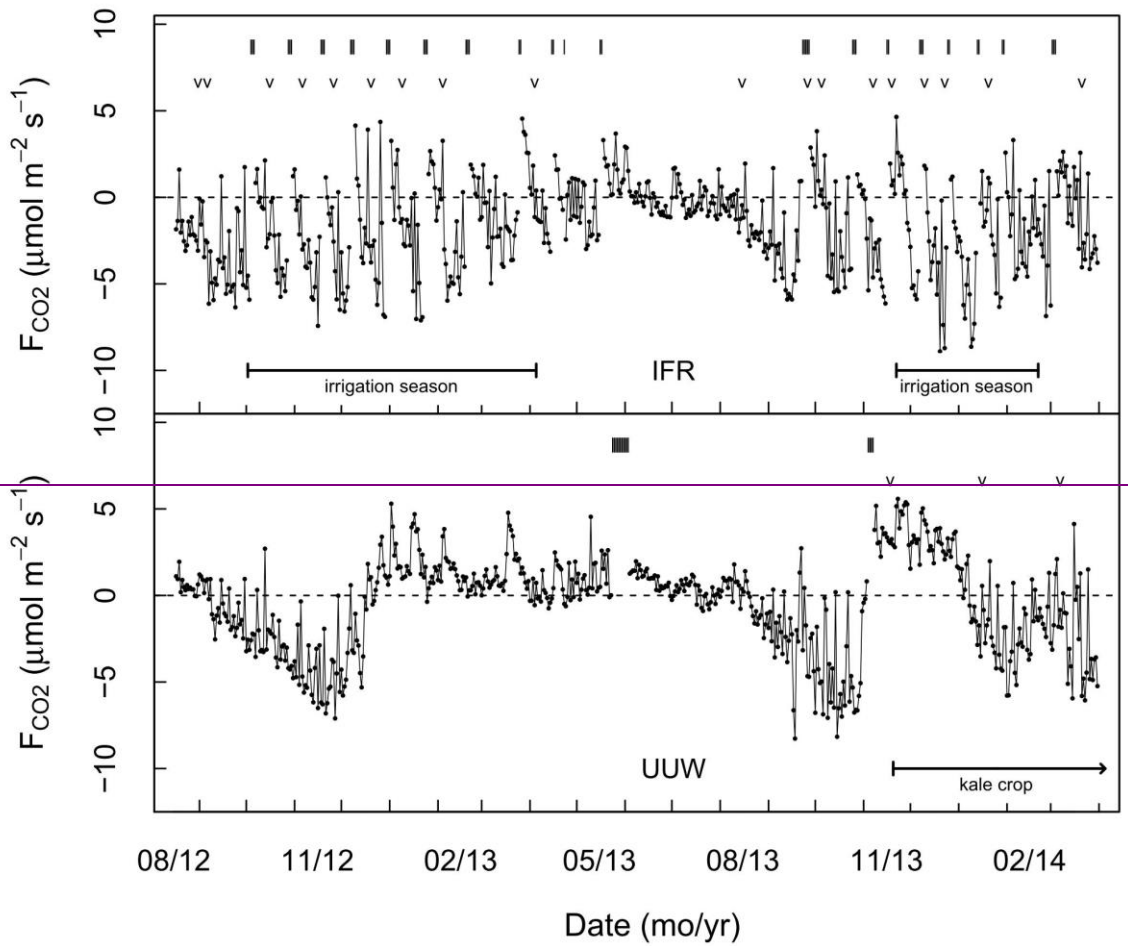
2

3 **Figure 2.** Left: evolution of CH₄, CO₂ and N₂O mole fractions (from top to bottom) at 0.76 m
 4 height, as well as standard deviation of vertical wind speed (σ_w), at the IFR site, for a few
 5 days. Horizontal short-dashed lines indicate the southern-hemisphere background values of
 6 the mole fractions. The horizontal long-dashed line marks the low-turbulence threshold for σ_w .
 7 Vertical dashed lines enclose an example period of low turbulence. Right: Linear regressions

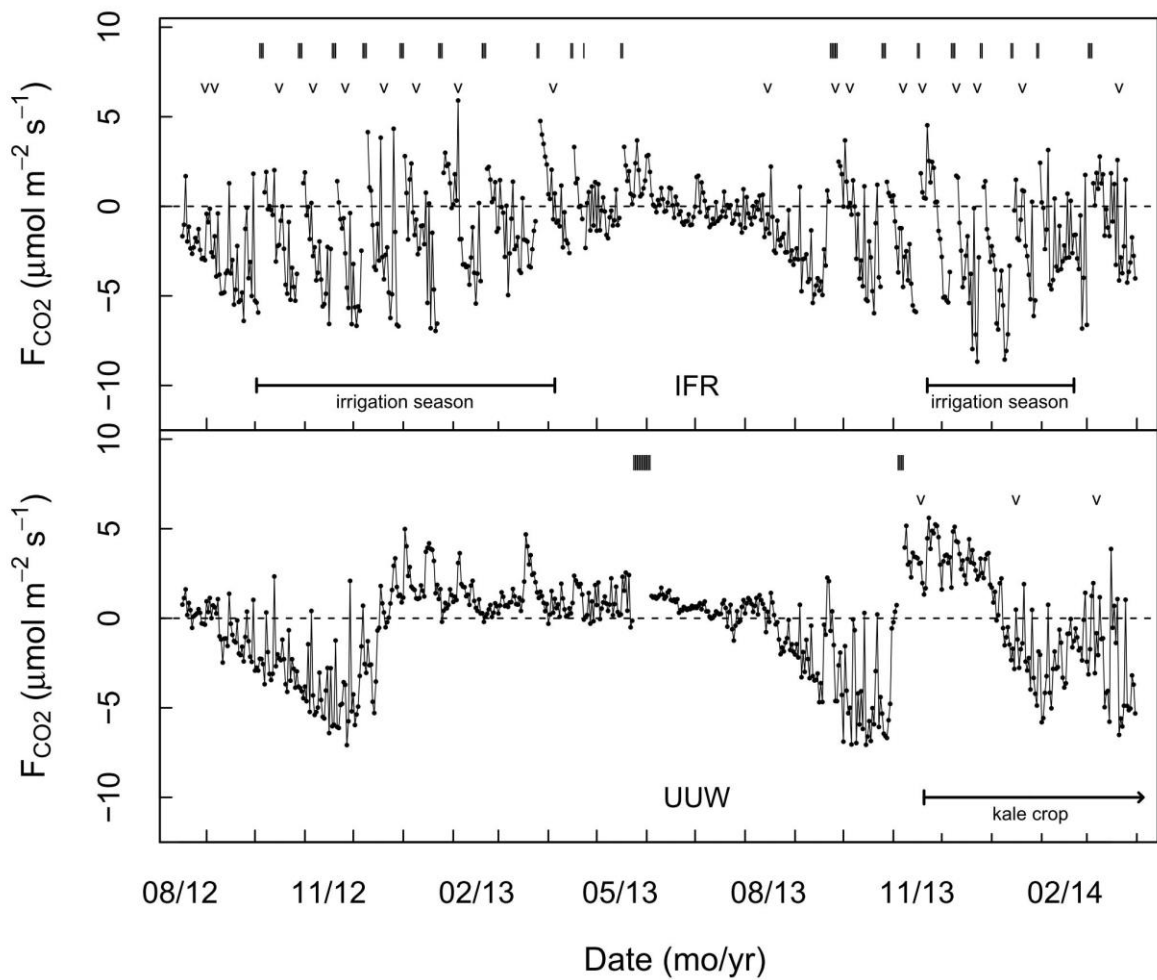
1 of mole fractions for CH₄ (top panel) and N₂O (bottom panel) vs that of CO₂, for this example
2 period.

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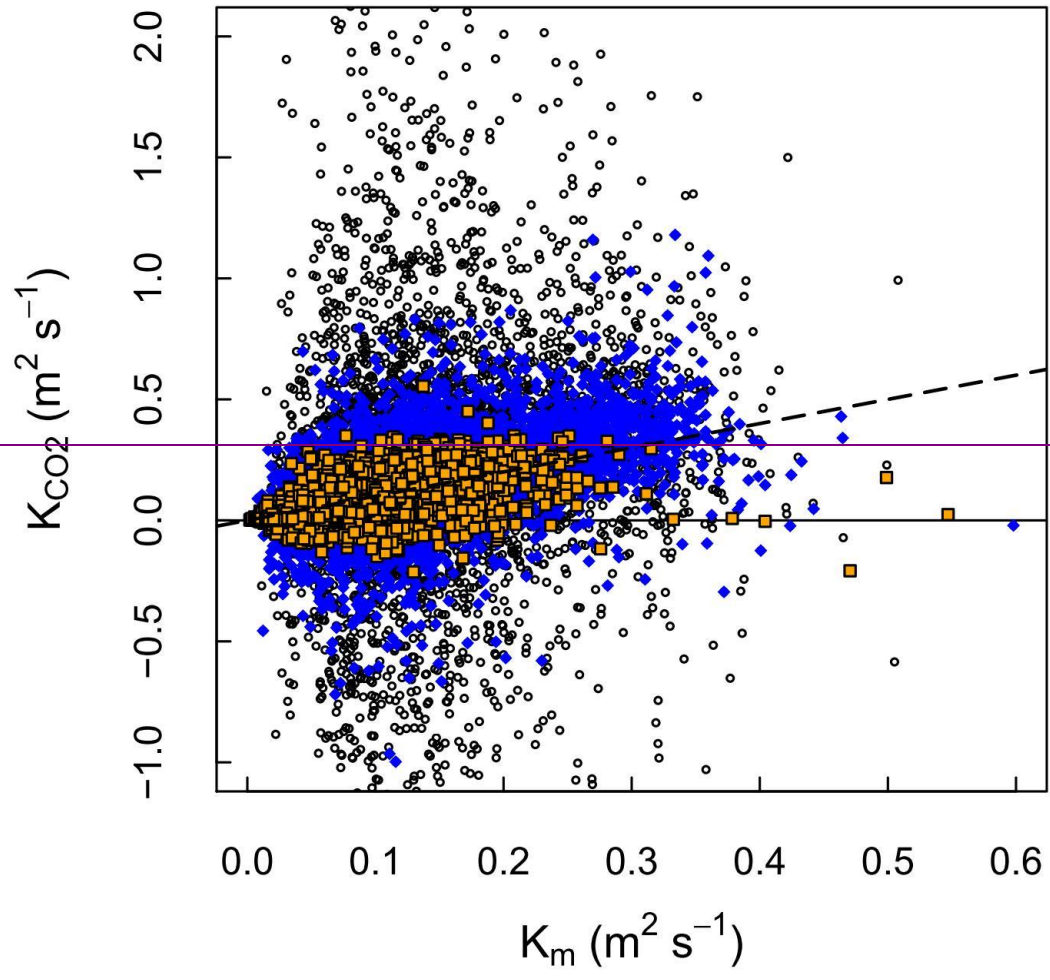


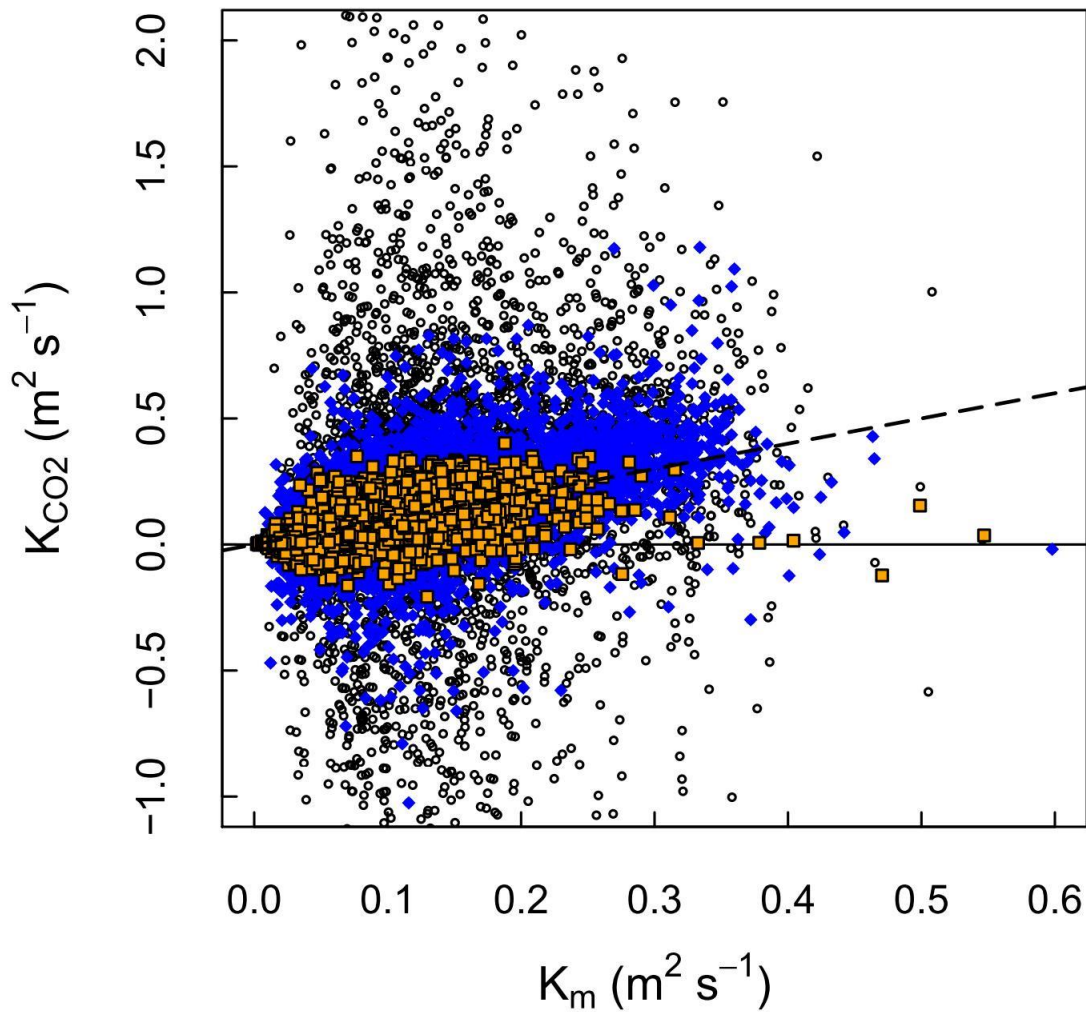
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2 **Figure 3.** Time series of the daily mean CO₂ flux (net ecosystem exchange) at the IFR site
 3 (top) and the UUW site (bottom). Grazing times are indicated by bars near the top of each
 4 panel. The times of fertiliser applications are marked by “v” symbols.

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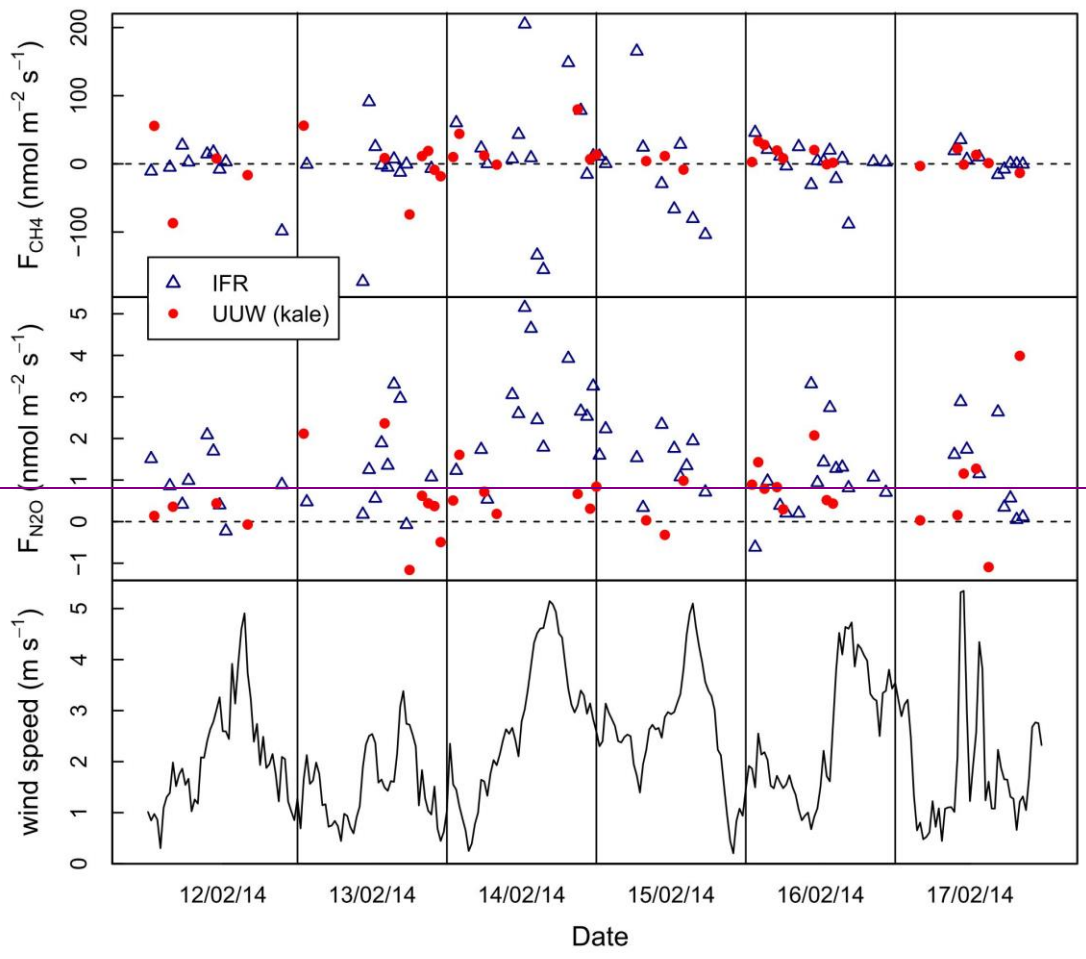


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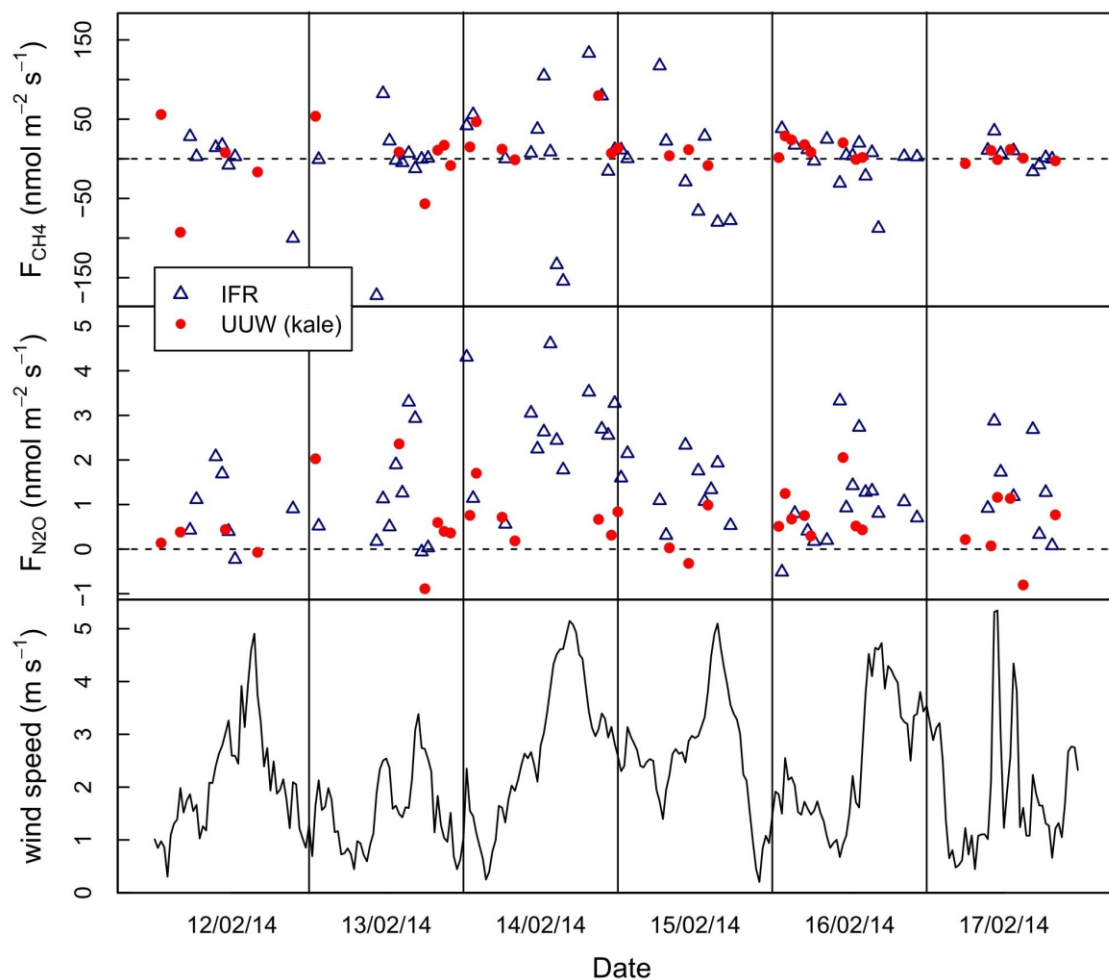
2 **Figure 4.** Half-hourly values of the turbulent diffusivity of CO₂ (from gradient measurements
 3 with the FTIR and flux measurements by eddy covariance) versus the diffusivity of
 4 momentum for neutral stratification (from sonic anemometer data), for the IFR site. Open
 5 circles: no filtering of $\Delta[CO_2]$ (mole-fraction difference between intake heights), solid
 6 diamonds: $\Delta[CO_2] < 0.6$ ppm excluded, framed squares: $\Delta[CO_2] < 2.4$ ppm excluded. The
 7 dashed line is the 1:1 line.

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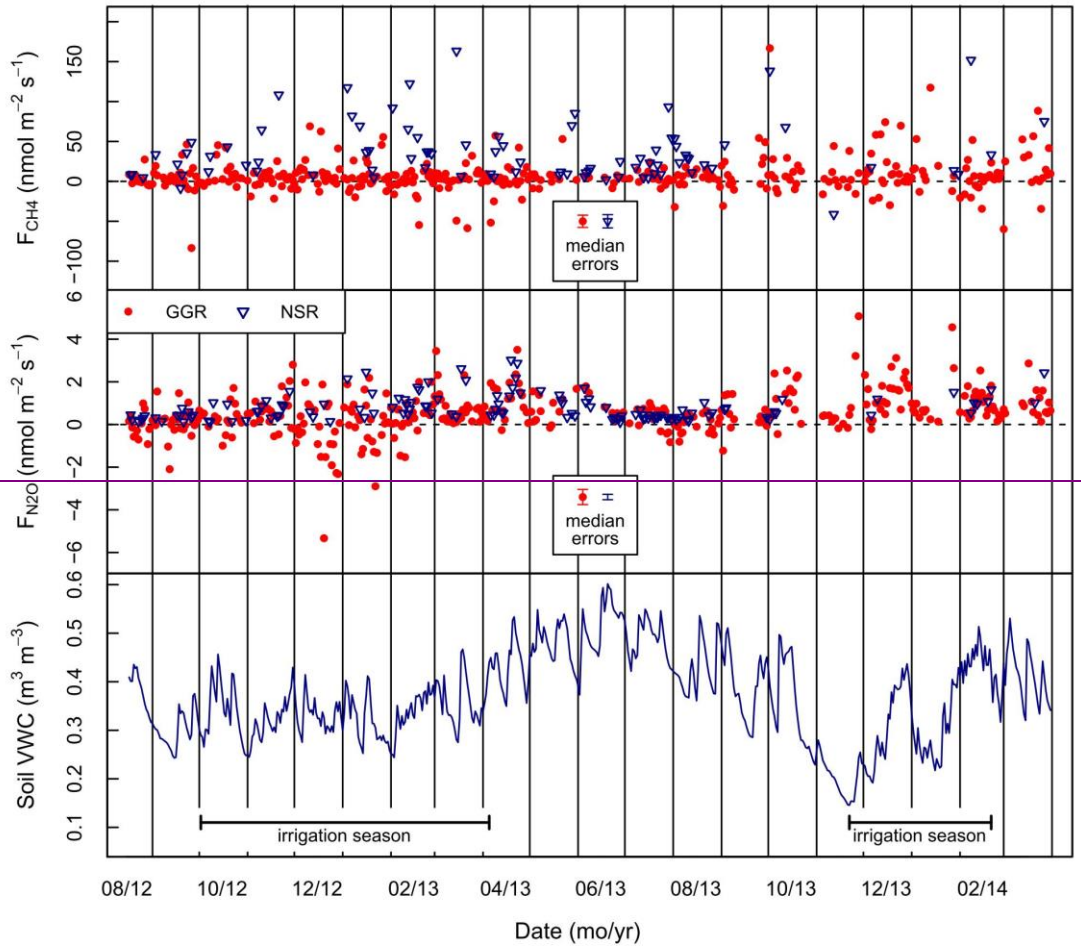


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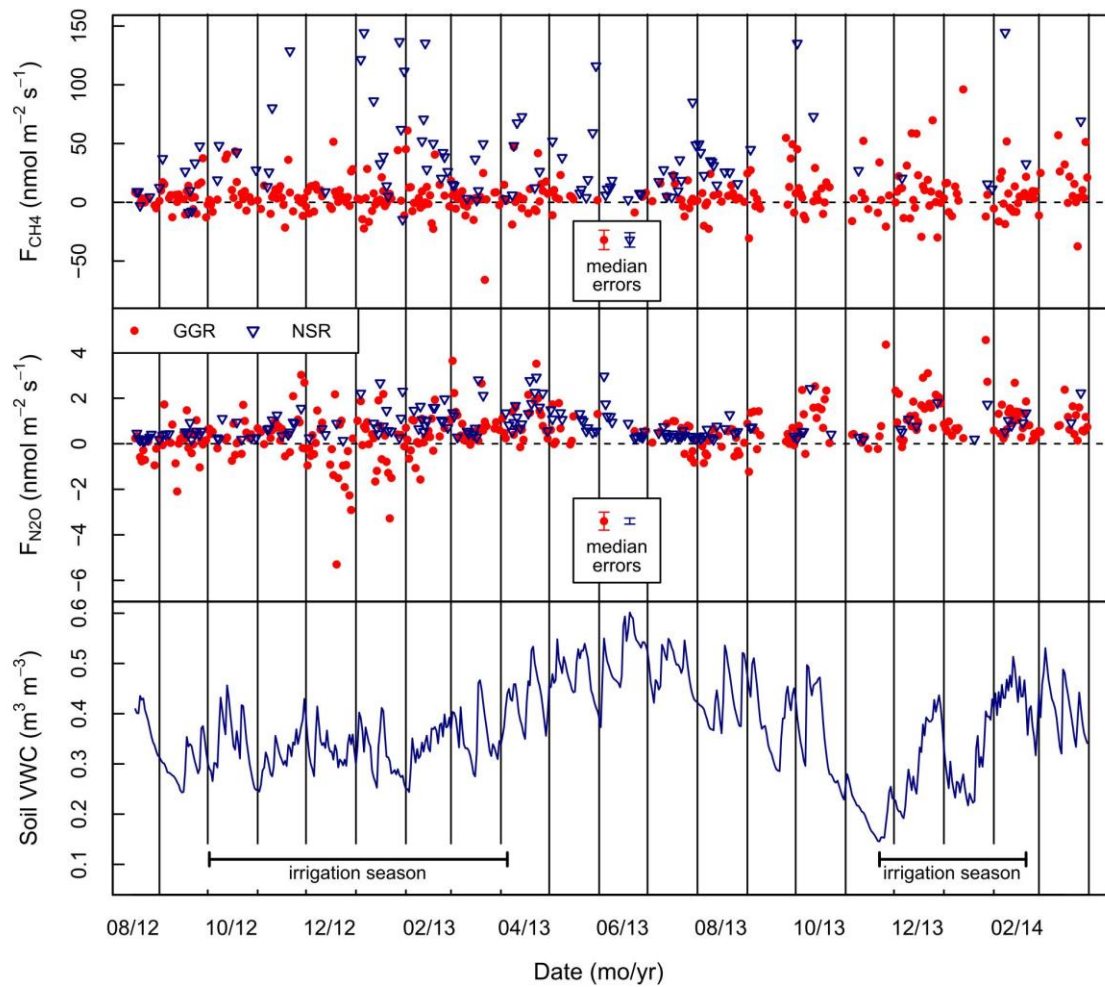


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 2 **Figure 5.** Half-hourly fluxes of CH₄ (top) and N₂O (middle) at the two sites, obtained with
 3 the GGR method, as well as wind speed (bottom), for an example period.

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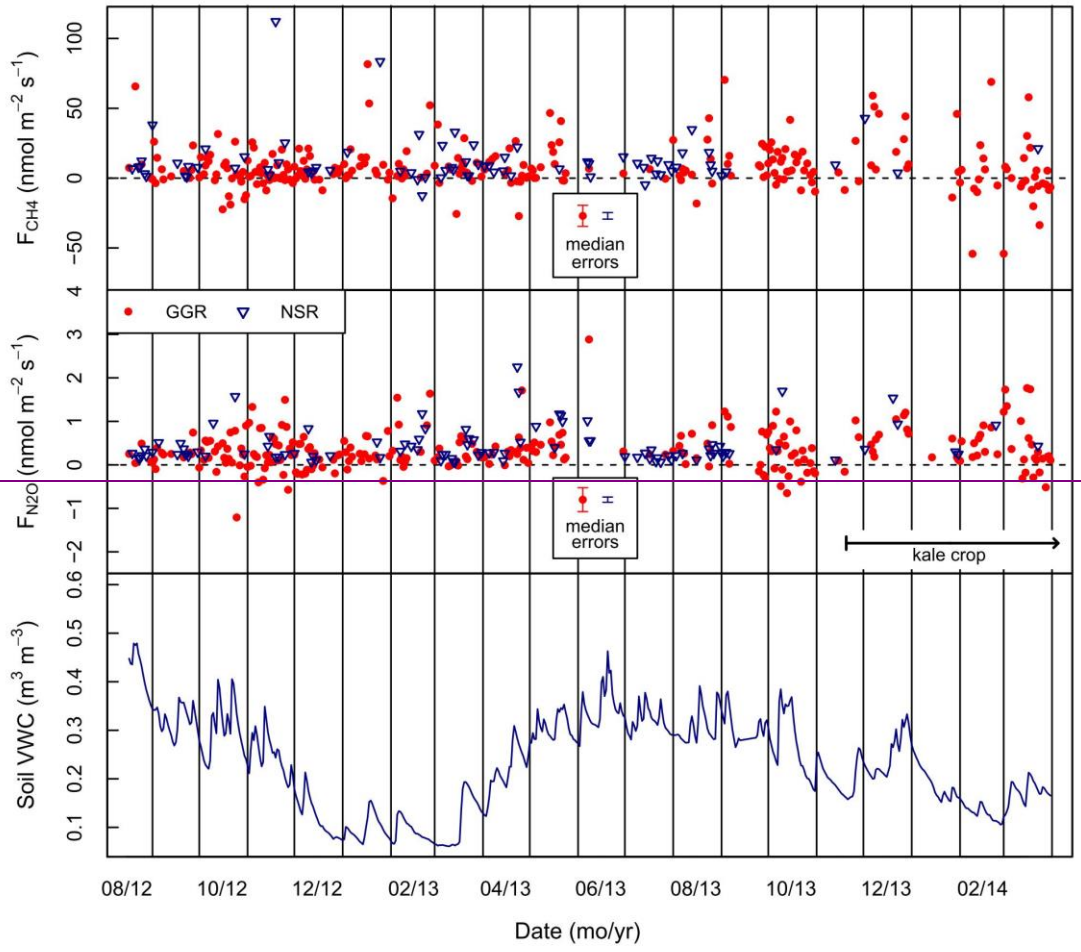
1

2 **Figure 6.** Daily mean fluxes of CH₄ (top) and N₂O (middle) at the IFR site, using the GGR
 3 method (dots) and using the NSR method (triangles). In these two panels, error bars in insets
 4 indicate for each method (GGR left, NSR right) the median of the standard error of the daily
 5 mean flux. Bottom panel: volumetric water contents at 5 cm depth.

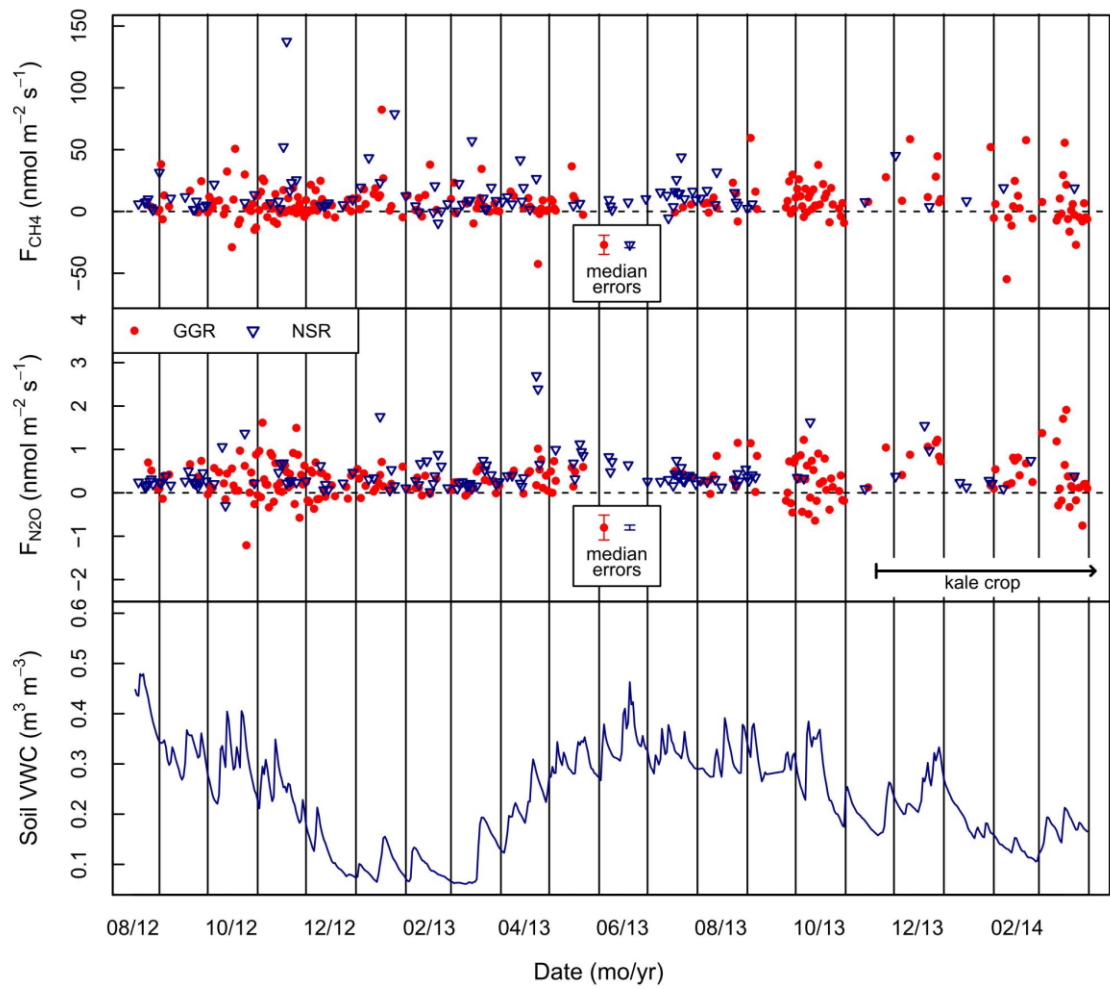
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7

8



1

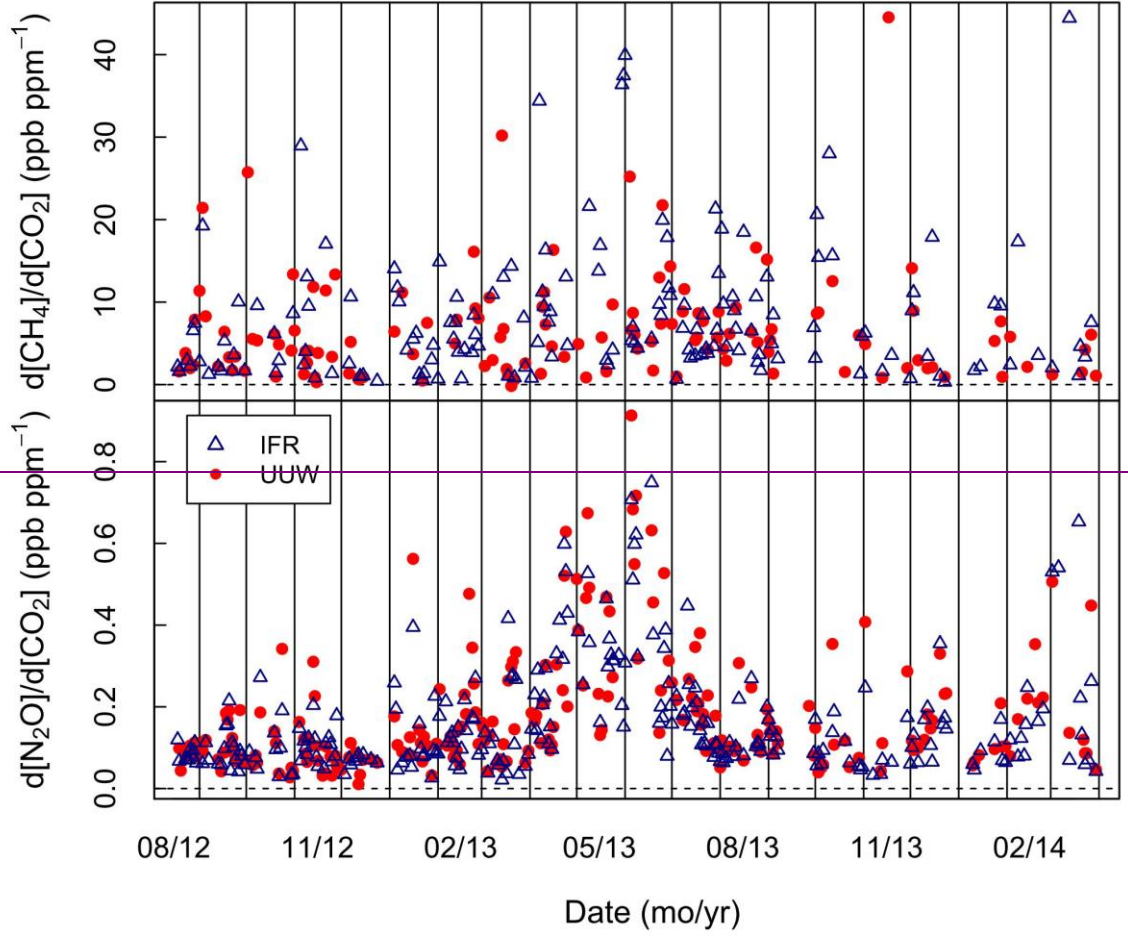


1

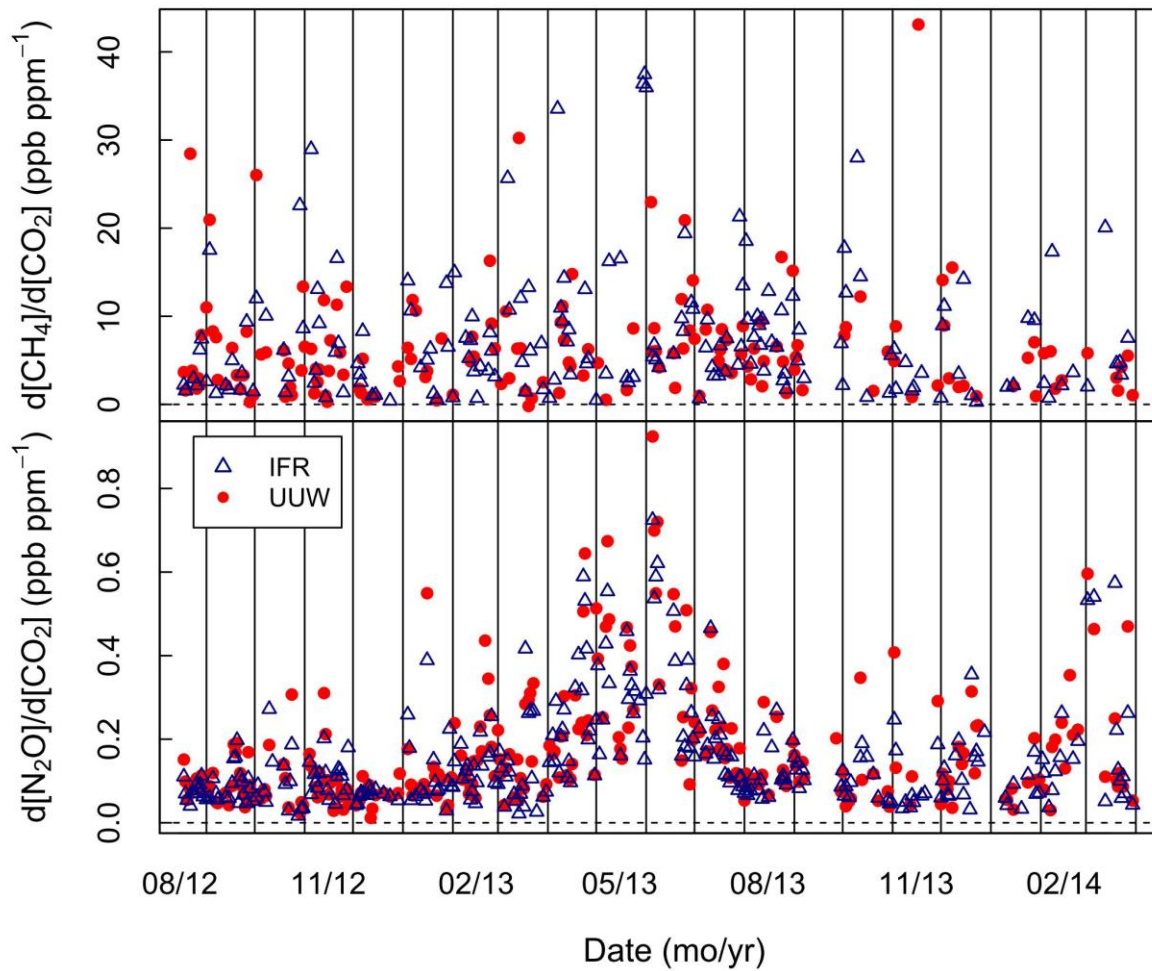
2 **Figure 7.** As Figure 6 but for the UUW site.

3

4



1



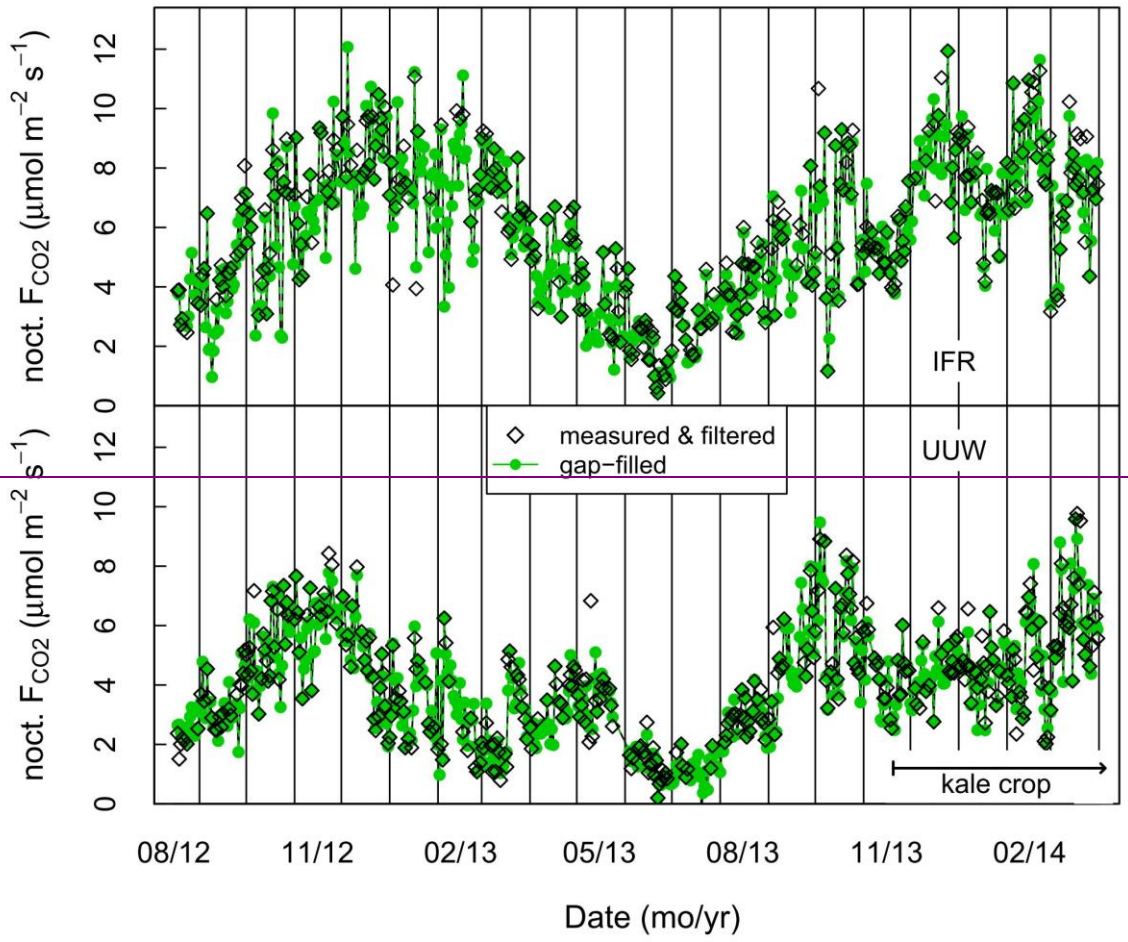
1

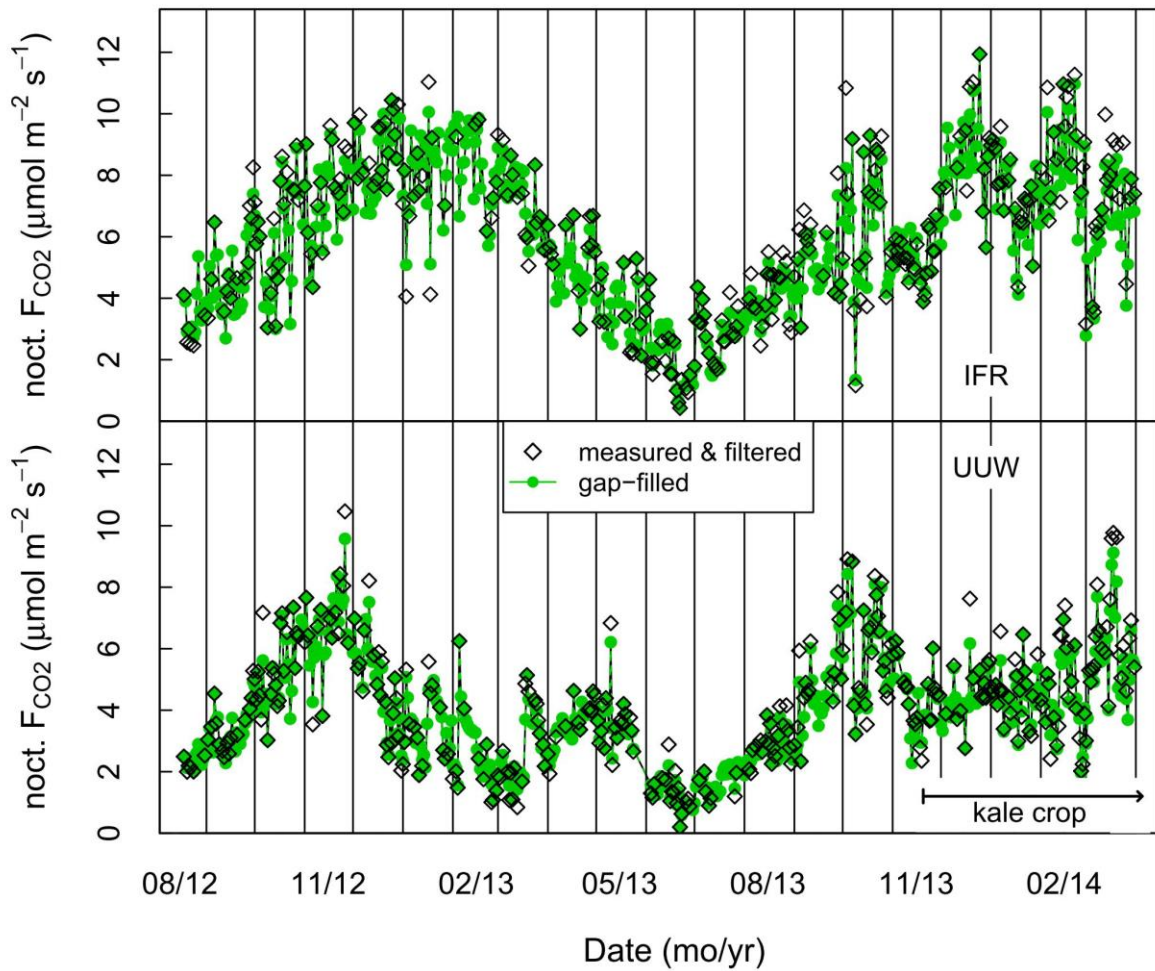
2 **Figure 8.** Temporal variation of the slopes of the linear regressions between CH₄ and CO₂
 3 (top panel) and between N₂O and CO₂ (bottom panel), for both sites.

4

5

6





1

2 **Figure 9.** Nocturnal CO₂ emission rates from the IFR site (top) and the UUW site (bottom).
 3 Each point is a whole-night average, obtained either only from measured values during
 4 periods with sufficient turbulence ($\sigma_w > 0.1012 \text{ m s}^{-1}$; open diamonds), or from the complete
 5 gap-filled dataset (solid dots).

6

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