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Low methane emissions from Australian estuaries influenced by geomorphology and disturbance

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Estuaries are a globally important source of methane, but little is known about Australia's contributions to global estuarine methane emissions. Here we present a first-order Australia-wide assessment of estuarine methane emissions, using methane concentrations from 47 estuaries scaled to 971 Australian estuaries based on geomorphic estuary types and disturbance classes. We estimate total mean (±standard error) estuary annual methane emissions for Australia of 30.56 ± 12.43 Gg CH_4 yr^{-1}. Estuarine geomorphology and disturbance interacted to control annual methane emissions through differences in water–air methane flux rates and surface area. Most of Australia's estuarine surface area (89.8%) has water–air methane fluxes lower than global means, contributing 80.3% of Australia's total mean annual estuarine methane emissions. Australia is a good analogue for the ~34% of global coastal regions classified as less than moderately disturbed (>40% intact), suggesting that these regions may also have lower methane fluxes. On this basis, recent global estuarine methane emission estimates that do not consider disturbance in their upscaling, probably overestimate global estuarine methane emissions.

Methane (CH_4) is a potent greenhouse gas with 27 or 80 times the global warming potential of carbon dioxide (CO_2) based on 100-year or 20-year time horizons[1]. Estuaries only cover 0.2% of global surface area[2] but may account for up to 2.7% of the mean annual CH_4 emissions[3] from global coastal and open oceans. However, early estimates of global mean CH_4 emissions ranging from 0.8 to 6.6 Tg CH_4 yr^{-1} included emissions from coastal wetlands[4] and/or highly disturbed European estuaries[4–7]. More recent estimates have reported lower global CH_4 emissions with a mean ± standard error of 0.90 ± 0.29 Tg CH_4 yr^{-1}[3] and a median of 0.23 (1st quartile to 3rd quartile: 0.02–0.91) Tg CH_4 yr^{-1}[3] and a median (1st quartile to 3rd quartile) of 0.25 (0.07–0.46) Tg CH_4 yr^{-1}[8]. These estimates incorporated more diverse estuary types, including those with lower emissions (e.g. fjords), and used improved (lower) global surface area estimates[2,9,10]. Although changes to catchment land cover and use, hydrology, and ecology (i.e. anthropogenic disturbance) may be an important control of estuarine CH_4 emissions, no previous studies have included the degree of estuary disturbance when upscaling to global CH_4 emissions. In addition, we know little about how geomorphology (estuary type) and anthropogenic disturbance interact in estuaries to influence CH_4 concentrations and water–air CH_4 emissions.

In estuaries, CH_4 is mostly produced during the microbial decomposition (methanogenesis) of estuarine- (autochthonous) and catchment-derived (allochthonous) organic matter[11]. Methanogenesis in estuaries occurs mainly in anoxic sediments[12] and is controlled by the availability of sulphate, organic matter, salinity, and oxygen in the sediments and/or benthic boundary layer[4]. As such, CH_4 concentrations generally follow a seaward decrease with increasing salinity, driven by a declining upstream supply of allochthonous organic matter and rising availability of marine-derived sulphate downstream (from <1 mmol l^{-1} of sulphate in freshwater up to 28 mmol l^{-1} in marine regions)[13]. Methanogenesis is also influenced by the level of anthropogenic disturbance (changes in land use and land cover, hydrology, and ecology) within estuaries and their catchments, which directly impacts the estuarine chemical, physical, and biological environment[14]. Land use changes associated with industrial, agricultural, and residential developments[14] impact estuarine water quality via increased pollutant inputs and runoff. Increased input of allochthonous organic matter and nutrients can stimulate autochthonous organic matter production[15–17] and enhance CH_4 production and emissions[18–20], with reports of wastewater inputs contributing up to 49% of estuarine CH_4 emissions[21]. Anthropogenic disturbances that alter estuarine biological and ecological characteristics include the introduction of invasive species, loss of native ecosystems, and extractive activities such as aquaculture, fishing, and water abstraction (e.g. construction of dams and sea walls)[14]. These disturbances can affect the availability of nutrients, plant density, and the tidal regime in the estuary[22], all of which may impact CH_4 production and

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emission. The combined effect of estuary disturbance is best accounted for using a 'holistic approach', where a water body is classified in an integrative manner that assesses biological, chemical, and physical characteristics as a whole rather than with a single objective quality metric[14].

Estuarine geomorphic features result from the interaction of factors such as the underlying geology, differences between river, wave, and tide energies, and channel basin and catchment characteristics (such as vegetation, climate, relief, soils, etc.)[23,24]. Categorising estuaries into geomorphic types is a useful tool for generalising hydrodynamic characteristics, such as depth, current velocity, tides, residence times, and stratification, which can be important controls on CH_4 emissions. For example, physical characteristics influence the distribution of intertidal environments[2] and affect water turbulence and the rate at which gas is transferred from water to the overlying air (i.e. the gas transfer velocity (k))[25–28]. These characteristics drive water–air exchange of CH_4 and therefore CH_4 emissions. Long water residence times driven by small tidal ranges (<3 m), coastal impoundment structures (e.g. weirs, breakwaters, coastal bars), and/or low current velocity in estuaries can enhance methanogenesis by increasing organic matter availability and decomposition[29,30]. If stratification occurs, the bottom layer is disconnected from atmospheric exchange and as a result, the anoxic bottom layer can have increased CH_4 concentrations but with a relatively lower overall CH_4 emission from the estuary[19,31]. In contrast, larger tidal ranges (>3 m) can increase CH_4 emission due to increased flushing of CH_4 from adjacent wetlands within the estuary (i.e. tidal pumping and lateral exchange)[32,33].

Globally, ~34% of coastal regions are classified as less than moderately disturbed (>40% intact)[34]. Australia is one of 21 nations where large expanses of relatively intact coastal regions (>60%) are found[34]. Australia has the 3rd lowest population density (3.3 people km^{-2}) globally[35], resulting in 75% of Australian estuaries being classified as low or moderately disturbed[36]. This makes Australia a good analogue for low and moderately-disturbed coastal regions globally. Australia's coastline measures 36,700 km, has 971 estuaries assessed for disturbance[36], accounts for 5.37% of global estuarine surface area[10], and has 1.82% of the global number of estuaries[37]. Despite Australia's contribution to global estuary number and surface area, and inclusion of low to moderate disturbance estuaries, CH_4 emissions have been measured for less than 2% of Australian estuaries (e.g.[21,38,39].), and only two of these estuaries were low or moderately disturbed. There is no Australia-wide estimate of CH_4 emissions from estuaries. In this study, we (1) estimate areal water–air CH_4 fluxes from 36 Australian estuaries and combine these with published CH_4 emissions from an additional 11 Australian estuaries[21,38] (total 47 estuaries, Fig. 1); (2) evaluate the influence of

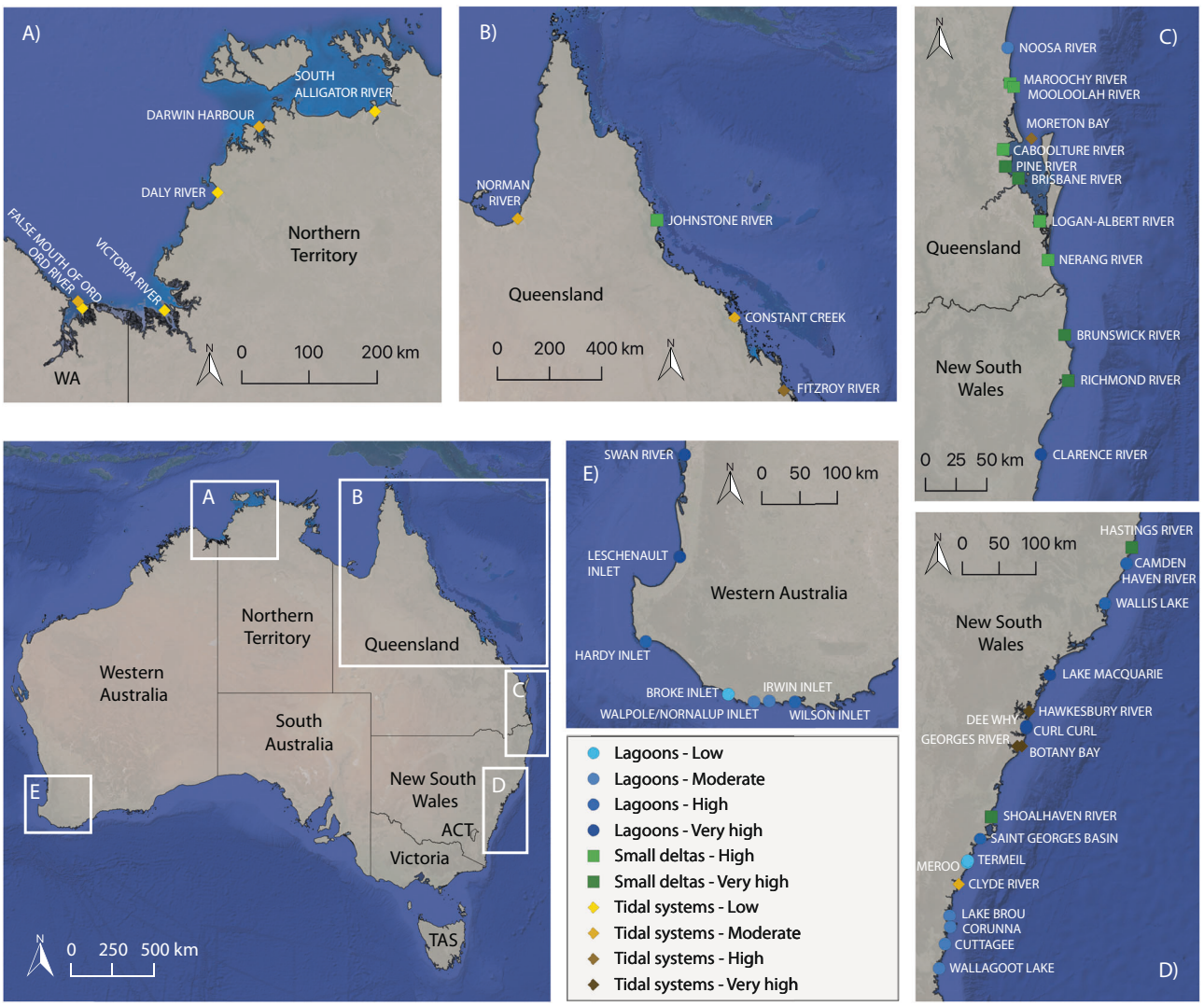

Fig. 1 | Map of study estuaries across Australia. The sample estuaries consist of 36 studied estuaries combined with three estuaries from Rosentreter et al.[38] and eight estuaries from Wells et al.[21] along the: **A** Northeastern Western Australia and Northwestern Northern Territory coast, **B** Northern Queensland coast, **C** Southern Queensland to Northern New South Wales coast, **D** New South Wales coast, and **E** Southwest Western Australia coast. Estuaries are categorised according to estuary type (represented by shapes) and disturbance class (indicated by colours)[36] (Base image ©Google Earth).

Table 1 | Total surface area coverage and estuaries represented by this study across Australia[40]

		Study surface area coverage			National surface area coverage		
Estuary type	Disturbance	(km^2)	Number of estuaries	% national representation	(km^2)	Number of estuaries	% of estuary type
Lagoon	Low	38	3	13.3	286	78	8.4
	Moderate	95	7	31.0	308	75	9.1
	High	226	5	32.1	704	82	20.8
	Very high	286	6	13.7	2083	36	61.6
	Not assessed	0	0	-	0	2	-
	Total	645	21	19.1	3382	273	8.6
Small delta	Low	0	0	0.0	99	38	16.7
	Moderate	0	0	0.0	85	39	14.4
	High	18	6	16.3	112	47	18.9
	Very high	100	6	34.0	295	25	49.9
	Not assessed	-	-	-	0	0	-
	Total	119	12	20.1	591	149	1.5
Tidal system	Low	1012	4	8.7	11598	359	32.7
	Moderate	1350	5	11.6	17152	97	48.4
	High	1553	2	13.4	5630	61	15.9
	Very high	179	3	1.5	582	27	1.6
	Not assessed	0	0	-	455	5	1.3
	Total	4094	14	11.6	35417	549	89.9
Total		4858	47	12.3	39390	971	100

The estuaries were classified into three geomorphic estuary types using morphological characteristics and four disturbance classes incorporating changes in catchment land use, ecology, morphology, and hydrology[40].

estuary type and disturbance[40] on CH_4 concentrations and emissions from these 47 estuaries; and (3) use geomorphic and disturbance classifications for 971 Australian estuaries[40] to scale CH_4 emissions from these 47 estuaries to the whole of Australia to estimate Australia's contribution to global estuarine CH_4 emissions. We hypothesise that both estuary type and the level of disturbance would significantly influence estuarine water CH_4 concentrations and water–air CH_4 fluxes, and that estuary type would interact with disturbance to influence total CH_4 emissions in Australia. We further hypothesise that CH_4 fluxes per unit area from Australian estuaries would be lower than global estuary CH_4 flux rates because of the generally lower disturbance found in estuaries in Australia. By focusing on three geomorphic estuary types (lagoons, small deltas, and tidal systems) and four levels of anthropogenic disturbance, the contribution of less disturbed estuaries to global CH_4 emissions can be better estimated, lowering uncertainties in global estimates.

Results

The results here represent data for in-water CH_4 concentrations and water–air CH_4 fluxes measured in 36 sampled estuaries, combined with the same data published for 11 additional estuaries[21,38] (details in 'Methods' section). The field campaign occurred over the 2017 to 2019 Australian summer seasons. The estuaries were grouped into three geomorphic types (lagoons, small deltas, and tidal systems) and into four levels of anthropogenic disturbance (low, moderate, high, and very high)[40]. Together, the estuaries consist of 21 lagoons, 12 small deltas, and 14 tidal systems across Australia (Table 1).

Influence of geomorphology and disturbance on CH_4

CH_4 concentrations and water–air CH_4 fluxes differed significantly ($p = 0.001$) between the three estuary types (Fig. 2a). Lagoons ($n = 751$) had the highest mean (± standard error (SE)) CH_4 concentration and water–air CH_4 flux (95.8 ± 3.5 nmol l^{-1} and 183.9 ± 8 μmol CH_4 m^{-2} d^{-1}) driven by higher maximum CH_4 concentration and water–air CH_4 flux (2196 nmol l^{-1} and 12,510 μmol CH_4 m^{-2} d^{-1}) compared to small deltas ($n = 719$; max: 277 nmol l^{-1} and 680 μmol CH_4 m^{-2} d^{-1}) and tidal systems ($n = 1138$; max: 559 nmol l^{-1} and 1263 μmol CH_4 m^{-2} d^{-1}) (Fig. 2a and Table 2). Lagoons also had the highest median concentrations and fluxes, indicating that the high mean water–air flux was not only due to the extremely high outliers (Table 2). Small deltas had the smallest range and the lowest maximum CH_4 concentration and water–air CH_4 flux (Fig. 2a and Table 2). Although small deltas and tidal systems had similar mean CH_4 concentrations, the mean water–air CH_4 flux was 16% higher in tidal systems compared to small deltas (Fig. 2a and Table 2).

Across all estuaries, higher disturbance significantly increased ($p = 0.001$) mean (± SE) CH_4 concentration (Fig. 2b1) and water–air CH_4 flux (Fig. 2b2) by approximately three times higher from the moderate disturbance group ($n = 633$; 28.2 ± 1.1 nmol l^{-1} and 66.4 ± 1.5 μmol CH_4 m^{-2} d^{-1}) to the very high disturbance group ($n = 888$; 85.1 ± 1.7 nmol l^{-1} and 191.0 ± 3.0 μmol CH_4 m^{-2} d^{-1}) (Table 2). However, low ($n = 356$) and high ($n = 731$) disturbance groups had similar CH_4 concentrations and water–air CH_4 fluxes ($p \le 0.202$). CH_4 concentrations and water–air CH_4 fluxes in the low and high disturbance groups were significantly higher compared to the moderate disturbance group ($p \le 0.002$) (Fig. 2b), which had the lowest CH_4 concentration and water–air flux (Table 2).

Between estuary types, the effect of disturbance on CH_4 concentration and water–air CH_4 flux differed, but was generally stronger in the higher (high and very high) disturbance groups (Fig. 3). CH_4 concentration in the low disturbance lagoons ($n = 41$) was significantly higher than in the moderate disturbance lagoons ($n = 161$; $p = 0.053$), but significantly lower than in high ($n = 261$) and very high disturbance lagoons ($n = 288$; $p \le 0.017$), for which CH_4 concentrations were similar ($p = 0.712$) (Fig. 3a1). The response of water–air CH_4 flux in lagoons to disturbance was slightly different from the response of CH_4 concentration. Water–air CH_4 fluxes in high and very high disturbance lagoons were significantly higher than in low and moderate disturbance lagoons ($p \le 0.016$), but water–air CH_4 fluxes were similar between the low and moderate disturbance lagoons ($p = 0.298$) (Fig. 3a2). The large mean water–air CH_4 flux in high-disturbance lagoons

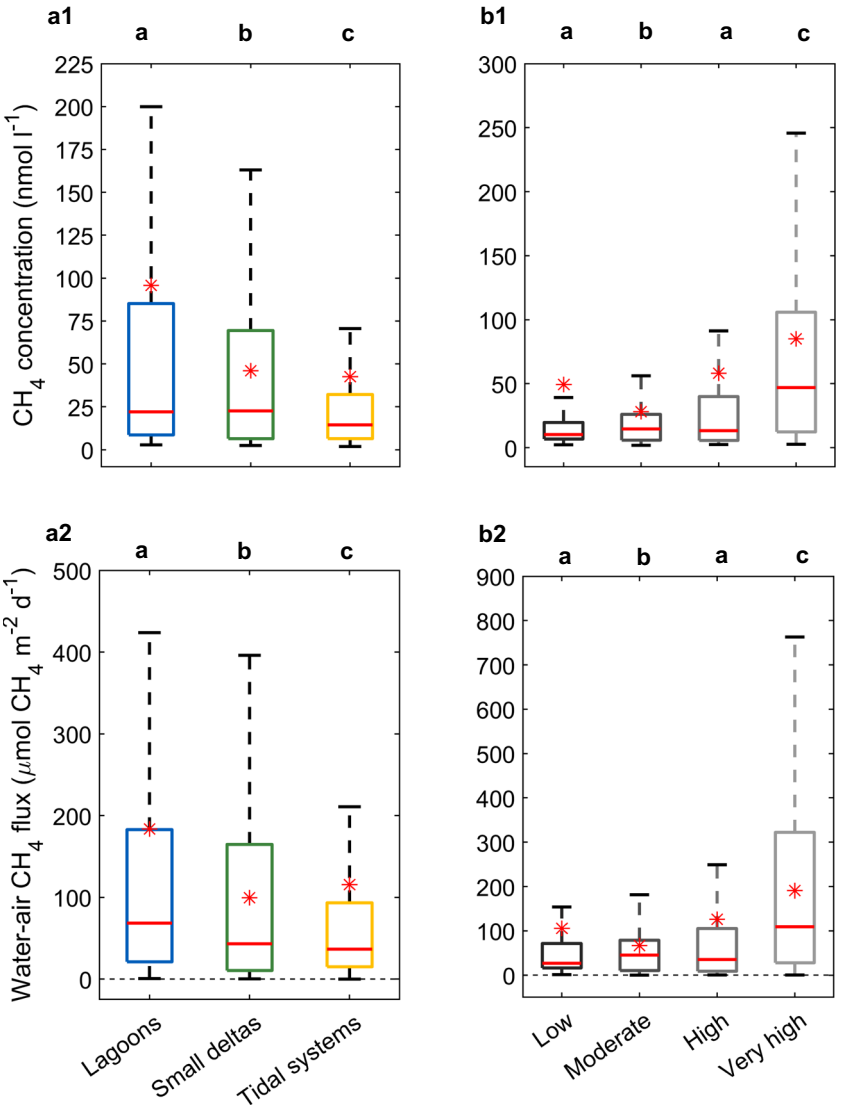

Fig. 2 | Methane concentrations and water–air fluxes in study estuaries grouped by estuary type and disturbance. Both CH_4 concentrations (row 1) and fluxes (row 2) are averaged per minute in estuary types: **a** lagoons (blue; $n = 3789$), small deltas (green; $n = 3622$), and tidal systems (yellow; $n = 5720$), and in disturbance classes (dark to light grey): **b** low ($n = 1796$), moderate ($n = 3189$), high ($n = 3677$) and very high ($n = 4469$). Each graph includes the mean (red asterisk), median (red bar), 1st and 3rd quartiles (box limits), and the furthest minimum and maximum values (whiskers) that fall within 1.5× the interquartile range. Outliers have been excluded from the graphs (Table 2). The letters above the plots signify statistical differences between groups. Dotted lines along the y-axis represent zero water–air CH_4 flux. Note the variations in y-axis scaling. Plot data can be found in Supplementary Data 1.

was driven by large outliers, as indicated by the lower median water–air flux for these systems (Fig. 3a2 and Table 2). In the moderate to very high disturbance lagoons, outliers of CH_4 concentrations and water–air CH_4 fluxes were larger than those in small deltas and tidal systems, regardless of the disturbance group. These large outliers in lagoons resulted in mean values that were higher than medians (Fig. 2a1, a2 and Table 2).

In small deltas, CH_4 concentration and water–air CH_4 flux significantly increased from the high ($n = 353$) to very high disturbance systems ($n = 366$; $p = 0.001$) (Fig. 3b). In tidal systems, only very high disturbance systems ($n = 234$) had significantly greater CH_4 concentration and water–air CH_4 flux compared to the other disturbance groups ($p = 0.001$; moderate group $n = 472$) (Fig. 3c). CH_4 concentration and water–air CH_4 flux in the low disturbance tidal systems ($n = 315$) were similar to those in high disturbance tidal systems ($n = 117$; $p \geq 0.444$) (Fig. 3c and Table 2). It should be noted that the minimum salinity measured in high-disturbance tidal system surveys was 21.6[40], which was associated with lower measured CH_4 concentrations and water–air CH_4 fluxes in these systems.

Effect of cleared catchment land on estuary CH_4

The mean percent of cleared catchment land increased with disturbance, from 10% in the low disturbance systems to 57% in very high disturbance systems (Table 2), and for lagoons was correlated with a significant increase in CH_4 concentrations ($n = 99$; partial correlations; $r = 0.415$ and $p = 0.001$) (Fig. 4a) and water–air CH_4 fluxes ($n = 99$; $r = 0.403$ and $p = 0.001$) (Fig. 4b). In tidal systems, CH_4 concentrations also significantly increased with percent cleared catchment land ($n = 103$; $r = 0.237$ and $p = 0.017$). In contrast, increases in percent cleared catchment land in small deltas were associated with decreased CH_4 concentrations ($n = 150$; partial correlations; $r = -0.34$ and $p = 0.001$) and water–air CH_4 fluxes ($n = 150$; $r = -0.22$ and $p = 0.007$), but no low or moderate disturbance systems were included, which limited the range of percent cleared catchments.

Seasonal differences in Australian estuarine CH_4 emissions

To assess winter water–air CO_2 fluxes, we calculated seasonal ratios using published summer and winter water–air CO_2 fluxes from 13 estuaries (Supplementary Table 1) and averaged them according to each estuary type. We subsequently applied these ratios to the summer water–air CO_2 fluxes observed in our current study to estimate their winter water–air CO_2 fluxes. Summer CH_4 water–air fluxes in lagoons and small deltas were higher than in winter (means: lagoons: 0.21 vs. 0.12 mmol CH_4 m^{-2} d^{-1} and small deltas: 0.09 vs. 0.08 mmol CH_4 m^{-2} d^{-1}, Table 3), with mean seasonal ratios of 0.59 in the lagoons and 0.87 in the small deltas (Supplementary Table 2). In contrast, tidal system winter water–air CH_4 fluxes were higher than in summer (mean: 0.28 vs 0.09 mmol CH_4 m^{-2} d^{-1}, Table 3) with a mean seasonal ratio of 3.02 (Supplementary Table 2). Overall, Australian estuaries in winter emit 0.16 ± 0.03 mmol CH_4 m^{-2} d^{-1} (mean ± SE), 10.6% higher

Table 2 | Statistics describing the percentage of cleared catchment land, and CH_4 concentration and water–air CH_4 fluxes calculated using per-minute resolution data in each estuary type (LA: Lagoons; SD: Small deltas; TS: Tidal systems), disturbance group (dist. group; 1: Low; 2: Moderate; 3: High; 4: Very high), and within the disturbance groups of the estuary types

		% cleared land	CH_4 concentrations (nmol l^{-1})						Water–air CH_4 flux (μmol CH_4 m^{-2} d^{-1})					
			Per-minute resolution (averaged per-estuary)						Per-minute resolution (averaged per-estuary)					
Estuary type	Dist. group	Mean (range)	Mean	Median	SE	IQR	Min	Max	Mean	Median	SE	IQR	Min	Max
LA	All	41 (0–100)	95.8	22.1	3.5	76.5	2.8 (42.9)	2196 (345)	183.9	69.6	8.0	161.7	0.6 (66.9)	12,510 (995)
SD	All	55 (27–79)	46.0	22.6	0.9	63.0	2.5 (3.8)	277 (142)	99.6	43.1	2.0	154.2	0.3 (3.8)	680 (362)
TS	All	29 (1–86)	42.5	14.5	1.0	25.7	1.9 (4.3)	559 (126)	115.5	36.6	2.5	78.3	0 (6.7)	1263 (308)
All	1	10 (0–31)	49.4	10.3	2.5	13.0	2.2 (21.7)	559 (123)	105.7	26.7	5.3	55.2	1.2 (37)	1263 (260)
All	2	24 (1–60)	28.2	14.6	1.1	20.1	1.9 (12.8)	670 (112)	66.4	45.3	1.5	68.4	0 (26.1)	534 (187)
All	3	54 (15–86)	58.2	13.3	3.1	34.2	2.5 (4.4)	2196 (295)	125.9	35.3	8.0	96.3	0.4 (7.6)	12,510 (1342)
All	4	57 (28–100)	85.1	46.9	1.7	93.5	2.6 (43)	981 (310)	191.0	109.2	3.0	294.2	0.3 (58.2)	1032 (536)
LA	1	18 (0–31)	56.9	33.0	3.5	68.8	9.4 (45.7)	158 (87)	115.7	84.3	4.0	87.1	50.2 (81.6)	232 (159)
	2	28 (5–50)	50.1	8.3	3.9	33.8	2.8 (18.2)	670 (128)	83.2	44.9	4.5	79.3	2.9 (37.5)	534 (146)
	3	43 (15–82)	109.9	18.4	8.3	89.9	3.1 (6.3)	2196 (598)	253.9	70.9	21.9	144.0	0.6 (15.8)	12,510 (3036)
	4	66 (39–100)	114.5	36.8	4.6	123.0	3 (100.8)	981 (515)	187.1	88.1	5.8	262.4	0.8 (136.4)	1032 (704)
SD	3	57 (27–79)	32.2	10.9	1.2	26.5	2.5 (3.2)	277 (124)	62.4	22.3	2.2	59.3	0.4 (2.8)	651 (334)
	4	52 (30–78)	59.2	48.9	1.3	66.2	2.6 (4.4)	258 (160)	135.4	101.0	3.0	191.8	0.3 (4.8)	680 (390)
TS	1	4 (1–5)	48.4	9.6	2.8	11.9	2.2 (3.7)	559 (150)	104.3	24.0	6.0	34.1	1.2 (3.5)	1263 (336)
	2	19 (1–60)	20.6	15.0	0.5	17.3	1.9 (5.2)	182 (91)	60.7	45.4	1.3	61.9	0 (10.2)	335 (244)
	3	73 (60–86)	21.0	7.9	1.0	25.4	2.8 (3.1)	98 (51)	32.2	12.0	1.9	32.1	1.1 (1.5)	253 (129)
	4	49 (28–62)	89.5	76.0	2.3	154.9	4 (4.4)	330 (202)	283.0	308.1	6.8	398.8	7 (8.6)	854 (494)

The mean per-estuary minimum and maximum CH_4 concentrations and water–air flux are provided in brackets.
SE standard error, *IQR* interquartile range.

than in summer (Table 3). Using water–air flux rates measured over only the summer season, Australian estuaries emitted an annual mean (± SE) of 18.17 ± 2.3 Gg CH_4 yr^{-1} at a rate of 0.14 ± 0.03 mmol CH_4 m^{-2} d^{-1} (Table 3), from a surface area of 39,390 km^2 (Table 1).

Tidal systems in summer contributed 69.8% of annual emissions, followed by 29.3% by lagoons and 0.9% by small deltas (Table 3). The higher winter CH_4 fluxes in tidal systems, combined with the large proportion of the total estuarine surface area (89.9%), resulted in larger winter estuarine CH_4 emissions in Australia (Table 3). The influence of tidal systems is reflected in annual CH_4 emission from Australian estuaries calculated using summer and winter CH_4 fluxes, which was 68.1% higher than annual CH_4 emission calculated using only summer CH_4 fluxes. This is despite summer and winter averaged CH_4 fluxes being only 5.3% higher than the summer CH_4 flux (Table 3).

Annual CH_4 emissions from all Australian estuaries

We estimate mean (± SE) annual CH_4 emissions (summer and winter) from Australian estuaries to be 30.56 ± 12.43 (range: 24.53–36.7) Gg CH_4 yr^{-1} (Table 3). Tidal systems accounted for the largest proportion of annual CH_4 emissions (85%), followed by lagoons (13.9%) and small deltas (1.1%). Interestingly, annual emissions of CH_4 were 4.9% lower in tidal systems, 0.4% lower in small deltas, and 5.3% higher in lagoons than what would be expected based solely on their surface area coverage (as indicated in Table 1). Annual CH_4 emissions from lagoons generally increased with increasing disturbance, whereas emissions generally decreased with increasing disturbance in tidal systems (Table 3). Mean (± SE) annual CH_4 emissions in Australian estuaries were highest in the moderately disturbed estuaries (10.4 ± 2.18 Gg CH_4 yr^{-1}) and lowest in the very high disturbance systems (3.99 ± 0.98 Gg CH_4 yr^{-1}). Of the total annual CH_4 emissions in Australia, 73.8% were emitted by low and moderately disturbed tidal systems (Table 3).

Discussion

There was a strong geomorphic control on CH_4 concentrations and water–air CH_4 fluxes in Australian estuaries. All geomorphic estuary types were annual net sources of CH_4 to the atmosphere, but of the three estuary types, lagoons had the highest CH_4 concentrations and water–air CH_4 fluxes (Fig. 2a). The long residence times characteristic of lagoons[41] due to low river inflow[37,42,43] and low tidal exchange[40] likely enhanced the trapping of autochthonous and allochthonous organic matter[4]. This is consistent with lagoons having the highest dissolved organic carbon (DOC) concentrations of all three estuary types[40]. Furthermore, the strongest inverse relationship between the water–air CH_4 fluxes and tidal range was found in lagoons, showing that water–air CH_4 fluxes increased as the tidal range[40] (Fig. 5b and Supplementary Notes) decreased (i.e. longer residence times).

Lagoons also had the largest range in CH_4 concentrations and water–air CH_4 fluxes. This high variability in lagoons can be partly attributed to very high maximums for both CH_4 concentration and water–air CH_4 fluxes in the upstream sections of the lagoons (Table 2). For example, although the mean per-estuary maximum CH_4 flux in lagoons was ~3 times larger than in small deltas and tidal systems, the highest CH_4 flux in lagoons was ~18 times greater than in small deltas and ~10 times greater than in tidal systems (Table 2). This indicates that although mean flux rates in the lagoon basin section were generally low, upstream lagoon riverine sections can have very high flux rates. The narrow upstream sections of lagoons have a higher density of terrestrial shoreline vegetation (visual field and Google Earth observations), which would increase allochthonous carbon input[44–46] and enhance CH_4 emissions when the organic matter is decomposed[4]. CH_4 emissions in upstream sections of lagoons are likely to be further enhanced by low marine intrusion, as supported by the inverse trend between tidal range and water–air CH_4 fluxes (Fig. 5b and Supplementary Notes). Low marine intrusion would limit the availability of marine-derived sulphate,

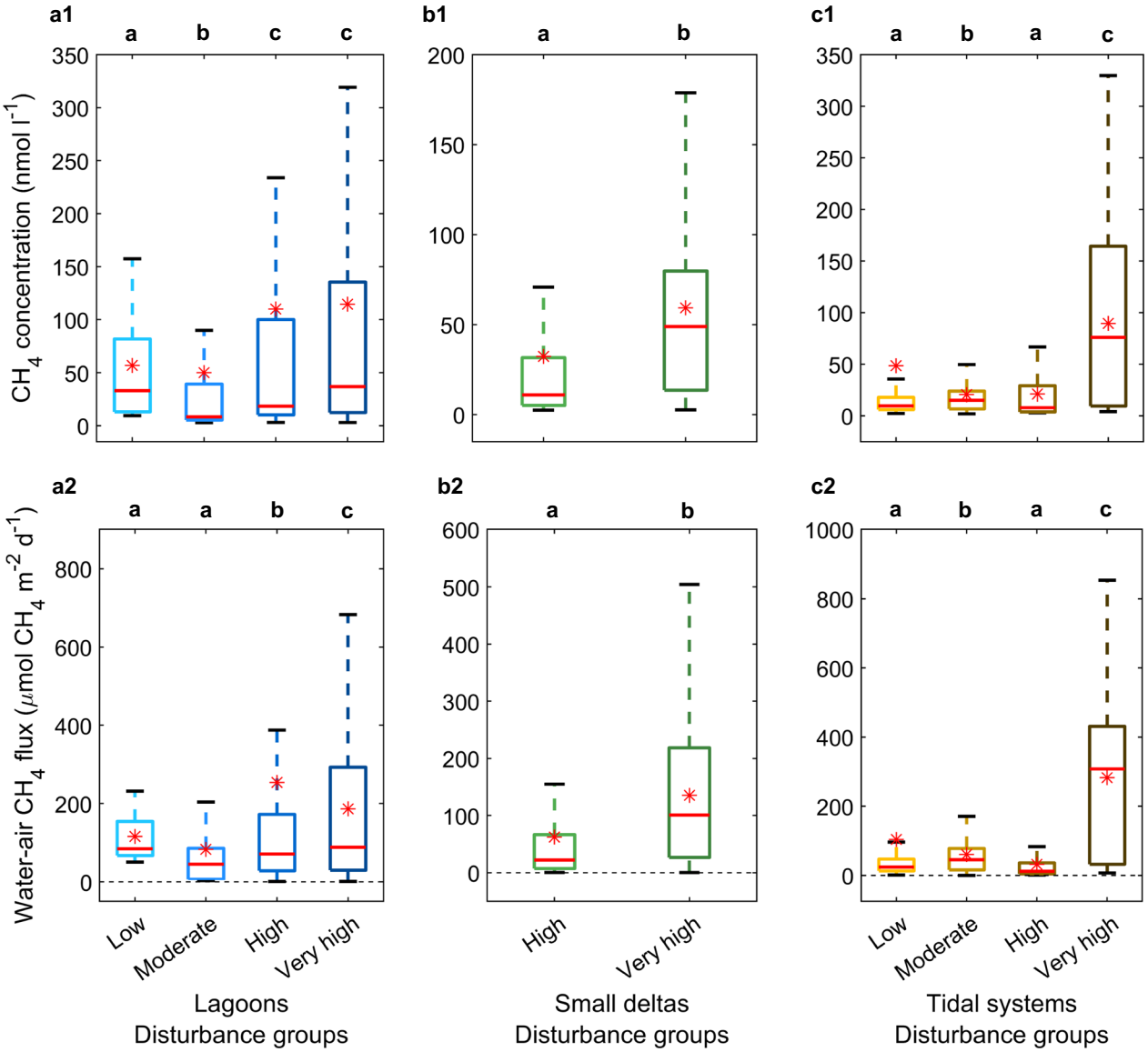

Fig. 3 | Methane concentrations and water–air fluxes in study estuaries grouped by disturbance classes within each estuary type. Both CH_4 concentrations (row 1) and fluxes (row 2) are averaged per minute across low to very high disturbance groups (light to dark colours)) in estuary type: **a** lagoons (blue; *n*: low = 214, moderate = 815, high = 1312, and very high = 1448), **b** small deltas (green; *n*: high = 1777 and very high = 1845), and **c** tidal systems (yellow; *n*: low = 1582, moderate = 2374, high = 588, and very high = 1176). Each graph includes the mean (red asterisk), median (red bar), 1st and 3rd quartiles (box limits), and the furthest minimum and maximum values (whiskers) that fall within 1.5× the interquartile range. Outliers have been excluded from the graphs. The letters above the plots signify statistical differences between groups. Dotted lines along the y-axis represent net-zero water–air CH_4 flux. Plot data can be found in Supplementary Data 1.

and reduce the competition between methanogens and sulphate-reducing bacteria, resulting in increased methanogenesis[4,47].

Overall CH_4 concentrations and water–air CH_4 fluxes in small deltas and tidal systems were lower than in lagoons likely due to shorter water residence times[41], resulting in less time for organic matter degradation. Decreasing DOC concentrations alongside increasing dissolved inorganic carbon (DIC) concentrations from lagoons to small deltas and tidal systems[40] likely reflected laterally imported DIC and CH_4 from shoreline habitats to estuarine waters via tidal pumping[32,48]. Shoreline habitats trap DOC and release DIC into the water column. Increased lateral export of CH_4 (and DIC) was likely related to the strong hydrological connectivity of tidal systems and deltas with adjacent mangroves and saltmarshes[49,50]. High lateral inputs are consistent with the positive relationship between tidal range[40] and CH_4 concentration and water–air CH_4 fluxes (Fig. 5c and Supplementary Notes) in the small deltas studied here, and in other small deltas[51] and tidal systems[52,53]. Mean CH_4 concentrations were similar between the small deltas and tidal systems, but mean water–air CH_4 fluxes in tidal systems were higher compared to small deltas (Fig. 2a). Higher water–air CH_4 fluxes in tidal systems are linked to higher gas transfer velocities[40] driven by faster water current velocities compared to other estuary types[40].

CH_4 concentrations and water–air CH_4 fluxes generally increased from the moderate, to high, to very high disturbance groups (Fig. 2b). Enhanced CH_4 concentrations and water–air CH_4 fluxes have been found in impacted estuaries with higher pollution inputs (e.g. wastewater)[39,54] and higher DOC concentrations[39,55]. Within estuary-type classes, CH_4 concentrations and water–air CH_4 fluxes generally increased in higher disturbance systems (Fig. 3). However, the direction of the relationship between DOC concentration and disturbance was estuary-type specific[40]. Estuary type also influenced the direction of relationships between percent cleared catchment land and CH_4 concentration and to water–air CH_4 flux (i.e. estuary type modified the effect of disturbance on CH_4 concentrations and water–air CH_4 fluxes).

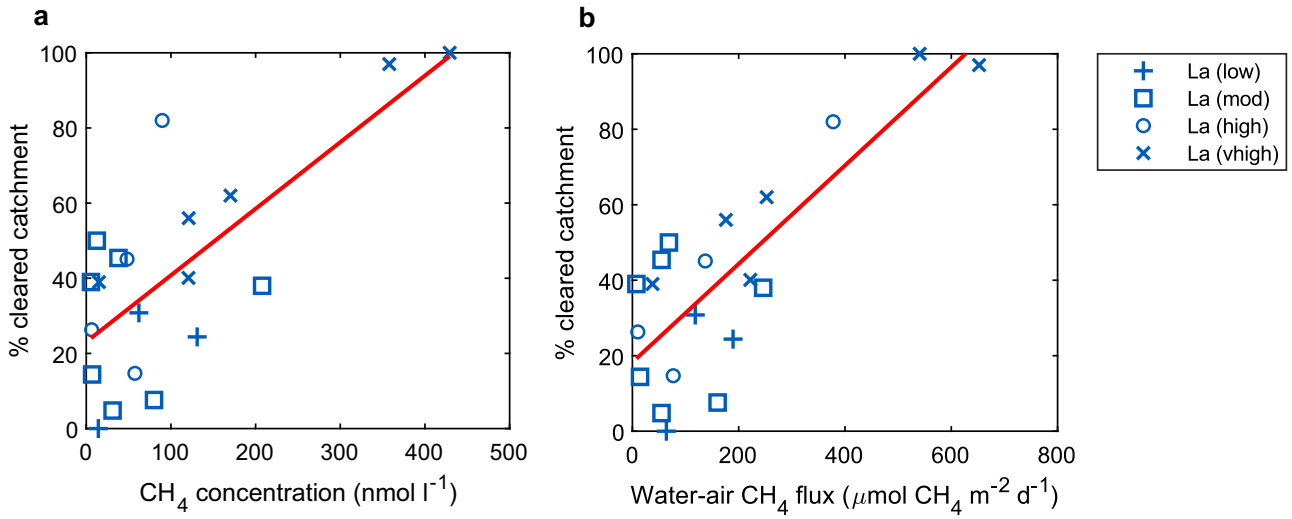

Fig. 4 | Relationships between percent cleared catchment land and CH_4 concentration and water–air flux in study lagoons. Linear fit line correlated between percent cleared catchment land and **a** mean CH_4 concentrations, and **b** mean water–air CH_4 flux in low ($n = 3$), moderate ($n = 7$), high ($n = 4$), and very high disturbance ($n = 6$) lagoons (La). Wilson Inlet was excluded as an extreme outlier (mean CH_4 flux of 1116 μmol CH_4 m^{-2} d^{-1} and land use change of 46%). Plot data can be found in Supplementary Data 1.

High and very high disturbance lagoons generally had higher CH_4 concentrations and water–air fluxes than low and moderate disturbance lagoons (Fig. 3a); CH_4 concentrations and water–air CH_4 fluxes increased with percent cleared catchment land (Fig. 4). DOC concentrations[40] were significantly correlated to CH_4 concentrations ($r = 0.29$ and $p = 0.004$) and water–air CH_4 fluxes ($r = 0.23$ and $p = 0.021$) in lagoons. Similar partial correlation was found for DOC concentrations and CH_4 concentrations ($r = 0.21$ and $p = 0.036$) or water–air CH_4 fluxes ($r = 0.2$ and $p = 0.043$) in tidal systems. However, high and very highly disturbed lagoons had generally lower DOC[40] than the low and moderate disturbance lagoons. These trends suggest that DOC concentrations did not drive increased CH_4 in higher disturbance lagoons. Rather, catchment land use changes, and associated variations in riverine input, likely affected organic matter quality and biogeochemical processes, including methanogenesis[56–58]. Long residence times in lagoons[2] have been shown to simultaneously enhance allochthonous organic matter degradation and autochthonous organic matter production[56], thereby enhancing CH_4 production.

Changes in CH_4 concentrations and water–air CH_4 fluxes with increasing disturbance in the tidal systems were less obvious and were only significantly higher in the very high disturbance systems compared to the lower disturbance systems (Fig. 3c). The less pronounced disturbance effect on CH_4 in tidal systems may reflect the larger tidal range[40] and associated larger water exchange compared to other estuary types, which in turn may mask any disturbance effect. However, low to highly disturbed tidal systems were all located in the largely undeveloped areas along the northern coastline[36], where catchment vegetative cover is high[36]. Therefore, low CH_4 concentrations and water–air CH_4 fluxes in the low to high-disturbance tidal systems could be due to naturally low allochthonous inputs from the catchments. In contrast, the very high disturbance tidal systems were located in heavily developed areas (i.e. Moreton Bay (Queensland), Port Philip Bay (Victoria), and Botany Bay (New South Wales)).

Australian estuaries overall emitted more CH_4 in winter than in summer. This was driven by relatively high winter CH_4 flux rates in tidal systems which account for 89.9% of Australian estuarine surface area. Lagoons and small deltas emitted less CH_4 in winter than in summer, but account for only 10.1% of estuarine surface area in Australia (Table 1). The impact of lagoons and small deltas on annual CH_4 emissions was therefore relatively low (Table 3). Seasonal differences in CH_4 emissions have not been extensively studied but a comparison of the Australian estuaries with estuaries in other countries suggests that seasonal trends differ between systems. For example, water–air CH_4 fluxes in the Rhine estuary in Germany were higher in the summer (median: 600 nmol l^{-1}) than in the winter (median: 310 nmol l^{-1})[7]. Similarly, summer water–air CH_4 fluxes were two times higher than winter fluxes in the tidal Tay estuary in the UK (summer: 0.35 g C m^{-2} yr^{-1}, winter: 0.15 g C m^{-2} yr^{-1})[59], and there were no seasonal differences in three other European tidal systems (Guadalquivir[60], Scheldt, and Gironde)[7]. Seasonal temperature differences have been proposed to influence CH_4, with higher microbial activity in warmer, summer months enhancing fluxes[61]. However, this is inconsistent with the observations in the current study, where overall fluxes were higher in winter. This may reflect less seasonal variation in temperatures in the tidal systems which are mostly in the tropics. Geomorphic factors (e.g. tidal influence) may have also influenced the seasonal differences in CH_4 emissions in tidal systems (and the other estuary types) such as changes in rainfall, lateral inputs, and biological activity, not just temperature. Shallower lagoons and small deltas may also be more sensitive to temperature changes due to a smaller buffering capacity compared to the larger tidal systems (Supplementary Notes). To account for seasonal water–air CH_4 flux variability[62], we applied the minimum and maximum seasonal ratios (Supplementary Table 2) to the summer flux rates to obtain a range of winter flux rates, which were used to calculate lower and higher estimates of annual CH_4 emissions (Table 3).

While we observed seasonal variability in Australian estuaries, the spatial variability within individual estuaries (overall mean of the minimum to mean of the maximum flux rate: 32.9–628.3 μmol CH_4 m^{-2} d^{-1}, difference of 595.4 μmol CH_4 m^{-2} d^{-1}) was substantially greater than seasonal variability (difference between the mean of summer and mean of winter estuary mean flux rates: 15.2 μmol CH_4 m^{-2} d^{-1}), indicating that spatial variability along salinity gradients is more important than temporal variability (i.e. summer and winter differences) in determining CH_4 emissions from Australian estuaries. The effect of salinity on CH_4 was supported by significantly decreased CH_4 concentration and water–air CH_4 flux with increasing salinity in all estuary types (Supplementary Notes and Supplementary Fig. 4). Episodic events may be an important source of variability[63,64], but were not captured in this study. Within each estuary-type class, the difference between the mean minimum rates of each estuary and mean maximum rates of each estuary in water–air CH_4 fluxes was 15-fold in lagoons (66.9 to 995 μmol CH_4 m^{-2} d^{-1}), 95-fold in small deltas (3.8–362 μmol CH_4 m^{-2} d^{-1}), and 46-fold in tidal systems (6.7–308 μmol CH_4 m^{-2} d^{-1}) (Table 2). Although the difference in minimum and maximum flux rates between lagoons had the smallest

Table 3 | Water–air CH_4 fluxes and total CH_4 emissions from Australian estuary types (LA: Lagoons; SD: Small deltas; TS: Tidal systems), disturbance groups (dist. group; 0: Not assessed; 1: Low; 2: Moderate; 3: High; 4: Very high), and within the disturbance groups of the estuary types

		Summer			Winter					Summer + winter (annual)									
		Water–air CH_4 flux ($mmol\ CH_4\ m^{-2}\ d^{-1}$)			Water–air CH_4 flux ($mmol\ CH_4\ m^{-2}\ d^{-1}$)					Water–air CH_4 flux ($mmol\ CH_4\ m^{-2}\ d^{-1}$)					Australian CH_4 emissions ($Gg\ CH_4\ yr^{-1}$)				
Est. type	Dist.	Median	Mean	SE	Median	Mean	SE	Mean (−)	Mean (+)	Median	Mean	SE	Mean (−)	Mean (+)	Median	Mean	SE	Mean (−)	Mean (+)
LA																			
	1	0.13	0.13	0.04	0.08	0.08	0.02	0.07	0.08	0.10	0.10	0.03	0.10	0.11	0.17	0.18	0.05	0.17	0.18
	2	0.06	0.09	0.04	0.03	0.05	0.02	0.05	0.06	0.04	0.07	0.03	0.07	0.08	0.08	0.13	0.05	0.13	0.14
	3	0.10	0.30	0.18	0.06	0.18	0.11	0.17	0.19	0.08	0.24	0.15	0.23	0.24	0.34	0.98	0.60	0.96	1.00
	4	0.18	0.30	0.10	0.11	0.18	0.06	0.17	0.19	0.14	0.24	0.08	0.24	0.25	1.72	2.95	1.00	2.89	3.01
	All	0.13	0.21	0.05	0.08	0.12	0.03	0.12	0.13	0.10	0.17	0.04	0.16	0.17	2.31	4.24	1.71	4.15	4.33
SD																			
	1[a]	-	-	-	-	-	-	-	-	-	-	-	-	-	0.05	0.05	-	0.04	0.08
	2[a]	-	-	-	-	-	-	-	-	-	-	-	-	-	0.04	0.04	-	0.03	0.07
	3	0.06	0.07	0.02	0.05	0.06	0.02	0.02	0.13	0.05	0.06	0.02	0.04	0.10	0.04	0.04	0.01	0.03	0.07
	4	0.12	0.12	0.02	0.10	0.10	0.02	0.04	0.25	0.11	0.11	0.02	0.08	0.18	0.19	0.19	0.03	0.14	0.32
	All	0.09	0.09	0.01	0.08	0.08	0.01	0.03	0.19	0.08	0.09	0.01	0.06	0.14	0.31	0.33	0.06	0.23	0.53
TS																			
	1	0.02	0.06	0.05	0.05	0.19	0.15	0.13	0.25	0.04	0.13	0.10	0.10	0.15	2.45	8.53	6.58	6.61	10.44
	2	0.06	0.07	0.01	0.17	0.21	0.03	0.15	0.27	0.11	0.14	0.02	0.11	0.17	11.43	14.03	2.00	10.87	17.19
	3	0.02	0.02	0.02	0.07	0.07	0.06	0.05	0.08	0.04	0.04	0.04	0.03	0.05	1.43	1.43	1.24	1.11	1.76
	4	0.28	0.22	0.10	0.85	0.66	0.30	0.46	0.86	0.57	0.44	0.20	0.34	0.54	1.94	1.51	0.69	1.17	1.85
	0[b]	-	-	-	-	-	-	-	-	-	-	-	-	-	0.28	0.50	0.16	0.39	0.61
	All	0.05	0.09	0.03	0.16	0.28	0.09	0.20	0.36	0.11	0.19	0.06	0.14	0.23	17.53	25.99	10.66	20.14	31.84
Australia		0.08	0.14	0.03	0.09	0.16	0.03	0.12	0.22	0.10	0.15	0.03	0.13	0.18	20.16	30.56	12.43	24.53	36.70
All	1	0.07	0.09	0.03	0.07	0.14	0.08	0.11	0.18	0.05	0.12	0.05	0.10	0.13	3.84	8.19	3.73	6.99	9.38
All	2	0.06	0.08	0.02	0.12	0.12	0.03	0.09	0.15	0.11	0.10	0.02	0.09	0.12	10.89	10.40	2.18	8.96	11.84
All	3	0.06	0.15	0.07	0.05	0.10	0.04	0.08	0.15	0.06	0.13	0.06	0.12	0.15	2.15	4.78	2.22	4.35	5.58
All	4	0.15	0.21	0.05	0.10	0.25	0.08	0.18	0.35	0.12	0.23	0.06	0.20	0.28	2.08	3.99	0.98	3.39	4.86

The descriptive statistics for summer water–air CH_4 fluxes were calculated using per-minute resolution data averaged per-estuary measured in this study, and winter water–air CH_4 fluxes were calculated based on seasonal ratios obtained by taking the difference between summer and winter fluxes (see methods). Up-adjusted means (+) using maximum seasonal ratios and down-adjusted means (-) using minimum seasonal ratios for CH_4 emissions and water–air CH_4 fluxes were obtained from the sensitivity analysis (Supplementary Table 2). *SE* standard error.

[a]The means and medians for annual CH_4 emissions from low and moderate disturbance small deltas were estimated as the mean of high and very high disturbance small delta values.

[b]Annual emissions from 'not assessed' tidal systems were calculated using mean tidal system flux rates.

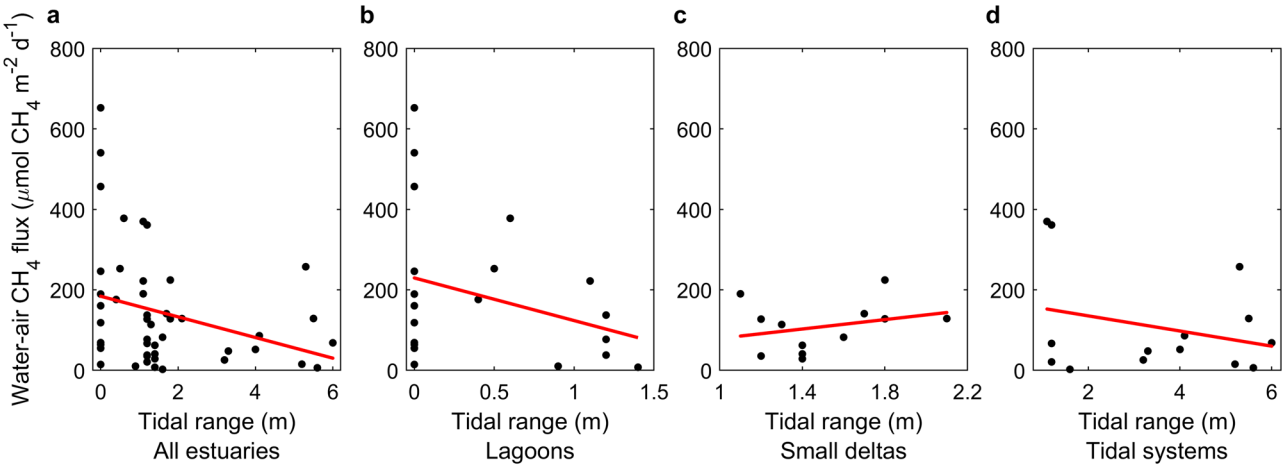

Fig. 5 | Relationships between estuary tidal range[40] and water–air CH_4 flux in all study estuaries and in estuary types. The linear fit lines indicate significant correlations (see Supplementary Notes). Data resolution is one sample per every 5 salinity change or 8 km travelled (discrete sampling intervals) across **a** all estuaries ($n = 47$), and in **b** lagoons ($n = 21$), **c** small deltas ($n = 12$), and **d** tidal systems ($n = 14$). Note the different scales on the x-axis. Plot data can be found in Supplementary Data 1.

magnitude of increase compared to the other two estuary types (15-fold compared to 95-fold and 46-fold), they had the highest minimum and maximum flux rates, and the largest range of flux rates (Table 2). This shows that the relationship between CH_4 flux rates and salinity depended on the estuary type and had a greater impact on overall estuary CH_4 emissions than seasonal variability. Therefore to capture the variations in CH_4 concentrations in estuaries, we used arithmetic means instead of medians. This approach allowed CH_4 concentrations and flux rates to be compared more effectively, incorporating the higher concentrations and flux rates in upper estuarine regions that would otherwise have been missed.

There are two recent compilations of global estuarine CH_4 emissions[3,8] which can be compared to the Australian estuarine CH_4 emissions estimated in the current study. Firstly, our mean (± SE) Australian estuarine emission was compared to the mean (± confidence interval) from Rosentreter et al.[3]. Because Rosentreter et al.[8] report median using a bootstrapping approach, their mean global estuary emissions were recalculated for tidal systems and deltas (mean 0.26 Tg CH_4 yr^{-1} using 0.15 mmol CH_4 m^{-2} d^{-1} and a global surface area of 294,956 km^2) and lagoons (mean 0.17 Tg CH_4 yr^{-1} using 0.16 mmol CH_4 m^{-2} d^{-1} and a global surface area of 179,946 km^2) for the purpose of better comparison with means in this study (Fig. 6a).

Mean water–air CH_4 fluxes in Australian low and moderately disturbed lagoons, high and very high disturbed small deltas, and low, moderately, and high disturbance tidal systems (Table 3) were lower than mean global estimates (all estuary types, 0.15 ± 0.02 mmol CH_4 m^{-2} d^{-1}) from ref. 3, and also lower than global mean fluxes in lagoons (0.16 ± 0.06 mmol CH_4 m^{-2} d^{-1}) and global mean fluxes in tidal systems and deltas (0.15 ± 0.03 mg CH_4 m^{-2} d^{-1}) from ref. 8. (Fig. 6a). This is significant because these Australian estuary disturbance classes account for 89.8% of the total estuarine surface area in Australia (Table 1 and Fig. 6b). Only the high (0.24 mmol CH_4 m^{-2} d^{-1}) and very high (0.24 mmol CH_4 m^{-2} d^{-1}) disturbed lagoons and very high tidal systems (0.44 mmol CH_4 m^{-2} d^{-1}, Table 3) had water–air fluxes greater than global water–air fluxes[3,8]. However, these systems make up only 8.6% of total estuarine surface area in Australia, and make only a small contribution (17.8%) to total estuary emissions in Australia (Fig. 6b). In addition, many of the global studies used an open water parameterisation (i.e. ref. 65). Had we only used the Wanninkhof[65] parameterisation, instead of an average of five parameterisations, our Australian estuarine CH_4 fluxes would have been even lower.

Lower water–air CH_4 fluxes in the majority of Australia estuaries compared to global fluxes probably reflect an overall lower catchment disturbance in Australia, which was captured in our sampling design and upscaling. Lower catchment disturbance is associated with low population density (3.3 people km^{-2})[35] and the delivery of lower riverine loads of carbon and nutrients[64,66] into Australian estuaries. In fact, Australia is part of ~34% of global coastal regions that are classified as less than moderately disturbed (>40% intact)[34]. Most of Australia's annual estuarine CH_4 emissions come from low and moderately disturbed tidal systems (73.8%, 22.55 Gg CH_4 yr^{-1}, Table 3) which are mostly found in remote northern Australia. This highlights the need to not only include a broad range of geomorphic types, but also for disturbance when upscaling estuarine CH_4 emissions.

Australia's estuaries have lower water–air CH_4 fluxes than estuaries globally due to lower disturbance, and total emissions are driven by low and moderately disturbed tidal systems in remote northern Australia. Assuming Australia is a good analogue for the ~34% of global coastal regions classified as less than moderately disturbed (>40% intact)[34], and given that recent global estuarine CH_4 emission estimates[3,8] do not include disturbance in their upscaling, global estuarine CH_4 emissions are probably overestimated. This highlights the need to include a broader range of geomorphic types and disturbance levels when measuring water–air CH_4 fluxes and upscaling regional and global estuarine CH_4 emissions.

Methods

Fieldwork description

36 estuaries were sampled for in-water CH_4 concentrations, physical parameters (water current velocity, water depth, and wind speed), and physicochemistry (salinity, temperature, pH, and dissolved oxygen) in the austral spring-summer season (2017–2019). The same data for 11 estuaries sampled under similar seasonal conditions were sourced from refs. 38,21, where the required parameters were available. The spatial surveys encompassed 21 estuaries along the New South Wales coastline (Merimbula to Brunswick Heads, Nov to Dec 2017), one in southeast Queensland (Moreton Bay, Oct 2018), seven along the north coast of Australia (Karumba, Queensland to Wyndham, Western Australia, Oct to Dec 2018), and seven in southwest Western Australia (Perth to Albany, Feb to Mar 2019) (Fig. 1 and Supplementary Table 3). Data for an additional eight estuaries in southeast Queensland were derived from ref. 21 (Sept to Oct 2014) and three in Queensland from ref. 38 (Oct 2016) (Fig. 1 and Supplementary Table 3).

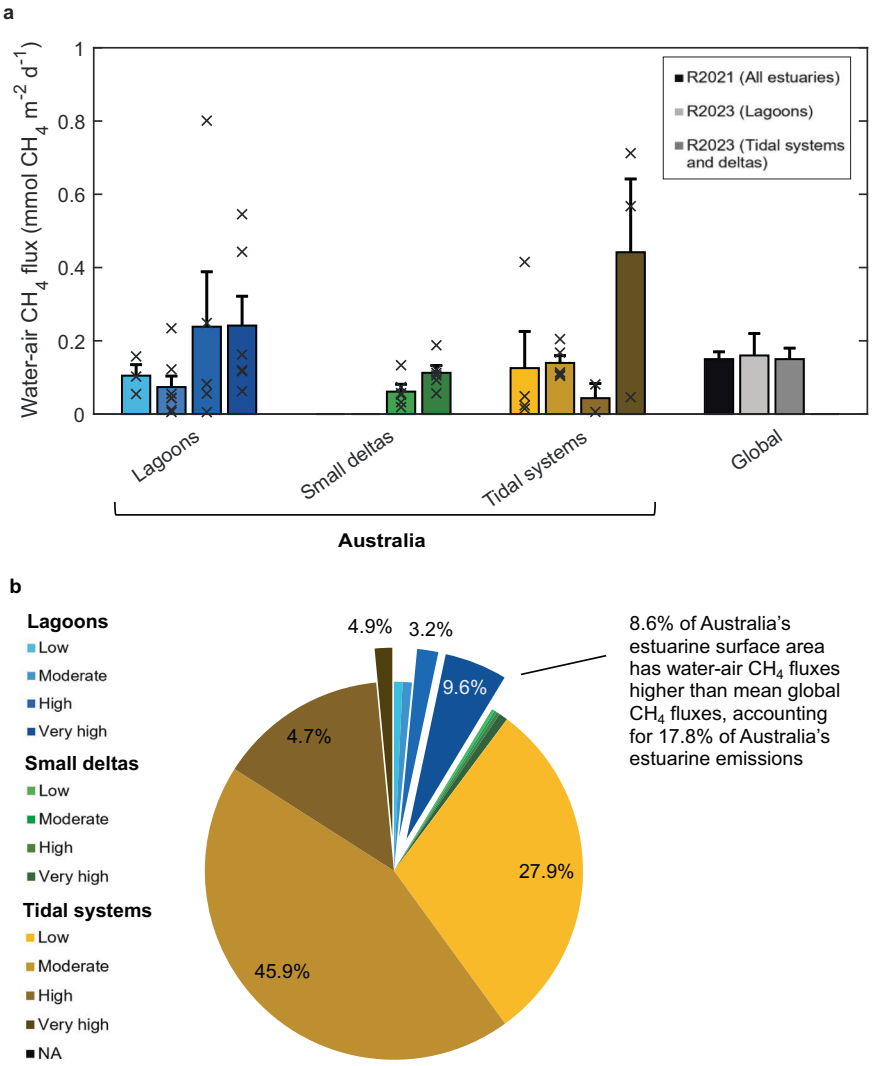

Fig. 6 | CH_4 flux rates and annual emissions in Australian estuaries compared to those globally. **a** Comparison of estuary mean (± standard error) water–air CH_4 fluxes in lagoons, small deltas, and tidal systems in Australia (this study; *n* presented in Table 1), with global mean water–air fluxes (all estuary types) from ref. 3 (R2021) and global mean water–air fluxes in lagoons, and in tidal systems and deltas from ref. 8 (R2023). **b** Relative surface area coverage of the different estuary types and disturbance classes in Australia shown by the sizes of the pie slices, and the percent contribution of each estuary type and disturbance class to total Australian CH_4 emissions shown by the % on each pie slice. *NA* not assessed (0% coverage).

Estuary classification schemes

Estuaries were selected to cover a large range of disturbance and geomorphic types according to the classifications of NLWRA[36] and ref. 2, respectively. NLWRA[36] assessed 971 Australian estuaries and described four disturbance classes (low (near-pristine), moderate (relatively unmodified), high (modified), and very high (extensively modified)), based on changes in catchment land use, estuary use, and ecology. The global estuarine typology of ref. 2 details three geomorphic types found in Australia: 1) lagoons (including Intermittently Closed or Open Lakes and Lagoons (ICOLLs) and estuaries with a central basin morphology), 2) small deltas, and 3) tidal systems (including drowned river valleys and tidal embayments) based on tidal influence, sedimentation, and hydrology. However, the existing classification of Australian estuaries[2] did not match our observations of satellite imagery, because it was developed with a low spatial resolution (0.5°, or 50 km). Therefore, we used the updated classification database by ref. 40, which categorised estuaries using the classification by ref. 2 combined with the estuarine disturbance database of NLWRA[36].

The distribution of estuary types across Australia corresponds to the tidal ranges along their respective coastlines (Supplementary Fig. 3). In northern Australia, macro-tidal regions are dominated by tidal systems while in the micro-tidal regions of southern Australia, lagoons are more prevalent. Our estuary selection includes all three estuary types with all four disturbance groups, except for low and moderate disturbance in small deltas (Table 1). The estuaries sampled and included in this study represent 12.5% of the total Australian estuarine surface area, consisting of 19.8% of lagoons, 10.3% of small deltas, and 11.8% of tidal systems in Australia (Table 1).

Underway data measurements

Boat-based survey transects were carried out in each estuary starting after high tide, from the marine mouth (~35 salinity) to the riverine-freshwater endmember (~2 salinity). Despite the aim of sampling a minimum salinity of 2, some estuary surveys were ended at higher salinities than 2 due to shallow depths and obstacles impeding further progress. The surveys were conducted in daylight hours, typically over a single day, with the larger estuaries requiring multiple days but no more than five days. A cruising speed of ~8 km h^{-1} was maintained whenever possible to ensure spatial and temporal consistency while the survey was underway. As a result, measurements taken in the estuary surveys reflect the spatial variation of parameters along the estuarine gradient (from marine to riverine). Underway physicochemical data and pCH_4 were measured via an integrated water-gas loop along the survey transect (Supplementary Fig. 1). Sample water was pumped from a depth of 0.5 m to 1 m using a 12 V pump with back-flow prevention (800GPH, Rule) and screen-filtered (High-flow filter basket, Ozito); before splitting into: (1) a flowthrough receptacle housing a physicochemical sonde (HL4, Hydrolab) recording per-minute measurements of temperature (±0.1 °C), salinity (±0.5%), pH_{NBS} (±0.2), and percent saturation dissolved oxygen ($DO_{\%sat}$, ±2%), and (2) a pair of interconnected showerhead exchangers (RAD Aqua, Durridge) where dissolved gases in the incoming sample water were equilibrated with the exchanger headspace,

and the dried air (Drierite) analysed in-line with a Picarro G−2508 cavity ring-down spectrometer (CRDS) measuring in-air CH_4 (ppmv)[67]. Corrections for water vapour pressure were applied to CH_4 dry mole fractions using methods of ref. 68, after which CH_4 dry mole fractions were converted into in-water CH_4 partial pressure (µatm) and concentration (nmol l^{-1}). The physicochemical sonde was calibrated regularly in the field, and the CRDS was serviced and calibrated by the manufacturer (±0.3 ppb) (Picarro, USA). Physicochemical data is presented in ref. 40.

Collection of discrete water samples, morphological, and meteorological data

Measurements for physical (water depth and current velocity) and meteorological data (barometric pressure, air temperature, and true wind speed) were collected concurrently with underway measurements (CH_4 concentrations and physicochemistry) at the start and end of surveys and at salinity intervals of 5. When salinity change was smaller than 5 per hour, samples were taken every hour instead (i.e. every 8 km travelled along the estuary) to account for changes in other estuarine parameters. Water samples for CH_4 were processed immediately after collection.

At each water sampling site, barometric pressure (±0.5 hPa @20 °C), true wind speed (±5% @10 m s^{-1}), and air temperature (±0.1 °C @20 °C) were measured using a vessel-mounted weather station (200WX, Airmar) 3 m above the water surface. Water depth was measured using a hull-mounted acoustic transducer (Airmar), and water current velocity was measured using a sub-surface drifter and the differential GPS-assisted Lagrangian method (adapted from Wetzel and Likens[69]). Current velocity measurements represented flow rates of the ebbing tide. In the smaller NSW lagoons, in-situ meteorological and water depth measurements were not collected. Instead, averaged daily meteorological measurements were obtained from the closest Bureau of Meteorology weather station[70] and averaged water depth data was taken from ref. 71. Physical and meteorological data are presented in ref. 40.

Water samples for CH_4 concentration were taken in the nine smaller, closed lagoons where the CRDS-equipped vessel could not be used for underway CH_4 measurements. Water samples for CH_4 analysis (6 ml) were syringe-filtered (0.22 µm PTFE Minisarts, Sartorius) at each sampling interval into duplicate 12 ml exetainers (Labco) pre-purged with N_2 with a needle through the septa[72], inverted, and refrigerated (4 °C) until analysis. The exetainers were loaded with 20 µl saturated $HgCl_2$ solution and pre-weighed before sample collection. The exetainers with samples were weighed again before analysis to determine the sample volume[21]. CH_4 concentrations in the headspace (coefficient of variation: ±5.2%) at room temperature were subsequently measured by gas chromatography (Shimadzu GC-9A) with a flame ionisation detector[21]. Measured gas-phase concentrations were converted into liquid-phase concentrations using the solubility calculations of ref. 73. CH_4 concentrations were linearly interpolated between sampling points into per-minute measurements along the survey transect to maintain spatial and temporal consistency between estuaries. Using interpolated CH_4 data, instead of averaged data, more accurately represented spatial gradients across lagoon basins.

The percentage of the estuary catchment cleared of natural vegetation (percent cleared catchment land) was based on existing data for New South Wales estuaries[71], southeast Queensland estuaries[74], Broke Inlet (Department of Water and Environmental Regulation (DWER), *unpublished data*), Irwin Inlet[75], Walpole Inlet[76], Hardy Inlet[77], Wilson Inlet (DWER, *unpublished data*), Swan River[78], Leschenault Inlet[79], Johnstone River[80], Constant Creek[81], Fitzroy River[82], Ord River estuaries[83], and for the other north Australian estuaries[36] (Supplementary Table 4). Mean tidal range data for the study estuaries were obtained from NLWRA[36].

Water–air CH_4 flux calculations

The per-minute water–air CH_4 flux (FCH_4; µmol CH_4 m^{-2} d^{-1}) was calculated using equation 1:

$$FCH_4 = kK_0\left(C_{water} - C_{air}\right)$$

where k is the gas transfer velocity of CH_4 (m d^{-1}) and K_0 is the CH_4 solubility coefficient (mol l^{-1} atm^{-1})[73] expressed as functions of temperature and salinity, and C_{water} and C_{air} are the partial pressure of CH_4 (µatm) in water and air, respectively[65]. Atmospheric CH_4 was taken to be the mean annual concentration of 1.86 ppm in 2018[84]. k_{600} was calculated based on five parameterisations obtained from the literature[25–27,65,85]:

$$k_{600} = -0.08 + 0.26v + 0.83u_{10} + 0.59h \quad (2)$$

Rosentreter et al.[27]

$$k_{600} = 1 + 1.719v^{0.5}h^{-0.5} + 2.58u_{10} \quad (3)$$

Borges et al.[25]

$$k_{600} = 0.314{u_{10}}^2 - 0.436u_{10} + 3.99 \quad (4)$$

Jiang et al.[85]

$$k_{600} = 0.77v^{0.5}h^{-0.5} + 0.266u_{10} \quad (5)$$

Ho et al.[26]

$$k = 0.251{u_{10}}^2\left(\frac{Sc}{660}\right)^{-0.5} \quad (6)$$

Wanninkhof[65].

In the first four parameterisations (Eqs. 2–5), k_{600} represents the gas transfer velocity normalised to Schmidt number of 600, v is water current velocity (cm h^{-1}), U_{10} is windspeed at a 10 m height (m s^{-1})[86], and h is water depth (m). The parameterisation by Wanninkhof[65] (Eq. 6) calculated for k to the Schmidt number at the measured temperature and salinity, which was then normalised to k_{600} using equation 7[65]:

$$k_{600} = k\left(\frac{600}{Sc}\right)^{-0.5}$$

where Sc was the Schmidt number and assuming a Schmidt exponent of −0.5 due to generally high water turbulences by tidal currents[87] found in estuaries. Because k was used to calculate the water–air flux of CH_4, k_{600} calculated using the other 4 parameterisations (Eqs. 2–5) were converted into k by rearranging equation 7. These parameterisations were chosen to reflect mangrove-dominated[26,27], tidal[25], lagoonal[85], and marine-dominated[65] environments found in estuaries. Although the parameterisation by Wanninkhof[65] was designed for marine open-water environments, we argue that it would be representative of the large, open tidal estuaries found in many tidal systems around Australia. Many estuary studies included in global estimates also include this parameterisation, making this current study comparable to other published estimates.

Water–air CH_4 fluxes in the eight southeast Queensland estuaries[21] were recalculated using the five parameterisations to ensure consistency. Recalculation was not possible for the three north Queensland estuaries[38] because water depth and current velocity data were unavailable. However, there was only one of these three estuaries in any given disturbance group and/or estuary type (i.e. moderate and high disturbance tidal system, and a high disturbance small delta, Table 1) and therefore, they should not introduce any systemic bias.

Data processing and statistics

Per-minute CH_4 concentration and water–air CH_4 flux calculations were reduced to 5-min averages to simplify data processing and analysis while maintaining high resolution and data features along the survey transects. Kolmogorov-Smirnov test for normality and Levene's tests for homoscedasticity returned significant results and together with an unbalanced study design, justified a non-parametric analysis approach. CH_4

concentrations and water–air CH_4 fluxes were not power-transformed but were normalised (z-score) before analysis. Not transforming the dataset retains the original heterogeneity of the mean-variance relationship and the spatial scale along the estuarine gradient, and avoids an inflated type I error as a result[88]. Differences in CH_4 concentrations and water–air CH_4 fluxes between estuary types (3 factors) and disturbance groups (4 factors) were analysed for significance ($\alpha = 0.05$; two-tailed) using the PERMANOVA procedure (Primer v7 with PERMANOVA+, PRIMER-e). Salinity was included as a covariate in the PERMANOVA analyses to account for possible effects on CH_4. The effect of salinity was confirmed by significant correlations between CH_4 and salinity (Pearson's correlation analysis, $\alpha = 0.05$, SPSS v25, IBM). Because PERMANOVA draws significance from tests based on permutations between datapoints, significant differences can be identified where descriptive statistics appear similar. 9999 permutations were executed using type I sum of squares with residuals under a reduced model. Significant differences between estuary types and the effect of disturbance within each estuary type were further investigated using pairwise tests (Primer v7 with PERMANOVA+, PRIMER-e). Correlations of CH_4 concentrations and water–air CH_4 fluxes with physicochemistry (temperature, pH, dissolved oxygen, and salinity[40]), percent cleared catchment land, tidal range, and DOC concentration[40] were power-transformed, normalised (z-score), and analysed using partial correlation ($\alpha = 0.05$) while controlling for salinity as a covariate (SPSS v25, IBM). Given that finding the drivers of CH_4 concentration and CH_4 emissions was the focus of this study, partial correlation analysis was chosen over exploratory multivariate methods such as Principal Component Analysis (PCA), allowing for targeted testing for correlations between variables and CH_4.

Upscaling estuarine CH_4 emissions to the Australian continent

Published summer and winter water–air CH_4 flux rates were available for ten[21,38] Australian small deltas and tidal systems. These seasonal CH_4 flux rates were used to calculate the seasonal ratios (winter:summer) for small deltas and tidal systems (Supplementary Table 1). The relationship between small delta CO_2[40] and CH_4 seasonal ratios ($S_{CH4} = 0.6106(S_{CO2}) + 0.2062$, Supplementary Fig. 2) was applied to estimate seasonal CH_4 ratios for lagoons (Supplementary Table 2). Indeed, the relationship between CO_2 and CH_4 for small deltas may not apply to lagoons in the same way. Therefore, the calculated seasonal ratios for lagoons should be interpreted with caution. After which, mean S_{CH4} were calculated for each estuary type and applied to summer water–air CH_4 fluxes to obtain winter flux rates of the 47 study estuaries (Table 3). The sensitivity of winter flux rates on annual CH_4 emission in Australian estuaries was also tested by applying the range of S_{CH4} to the annual CH_4 emission upscaling exercise (Table 3)[62]. The sensitivity range is reported alongside the overall annual CH_4 emission estimate.

Mean and median annual CH_4 emissions and their uncertainties (standard error) from all Australian estuaries ($n = 971$; Supplementary Fig. 3) were calculated by scaling disturbance-specific mean water–air CH_4 fluxes (μmol CH_4 m^{-2} d^{-1}) averaged per-estuary into the mean estuary type flux rate from the 47 study estuaries, and applied across the relevant total Australian estuarine surface areas (Table 1) of these geomorphic and disturbance types (area-weighting). Small deltas with low to moderate disturbance were not available for this study. The exclusion of these estuary types would result in an overestimation of CH_4 emissions from small deltas, but the impact is likely to be negligible, given that low and moderate disturbance small deltas account for only 0.5% of Australian estuarine surface area (all small deltas account for 1.5%). The mean and median small delta CH_4 flux were used in their place when upscaling annual CH_4 emissions.

Reporting summary

Further information on research design is available in the Nature Portfolio Reporting Summary linked to this article.

Data availability

The methane data measured and used in this study can be found in the data repository FigShare under the accession code https://doi.org/10.6084/m9.figshare.25933060. Source data for figures can be found in Supplementary Data 1.

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Author contributions

All authors have agreed to be listed and have approved the submitted version of the manuscript. J.Y. conceived the project, collected data, ran data analysis and interpretation, and led the writing of the manuscript. J.A.R. collected data and helped with data analysis, interpretation, and writing of the manuscript. J.O. collected data, contributed to interpretation, and helped write the manuscript. B.E. conceived the project, collected data, contributed to interpretation, and helped write the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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