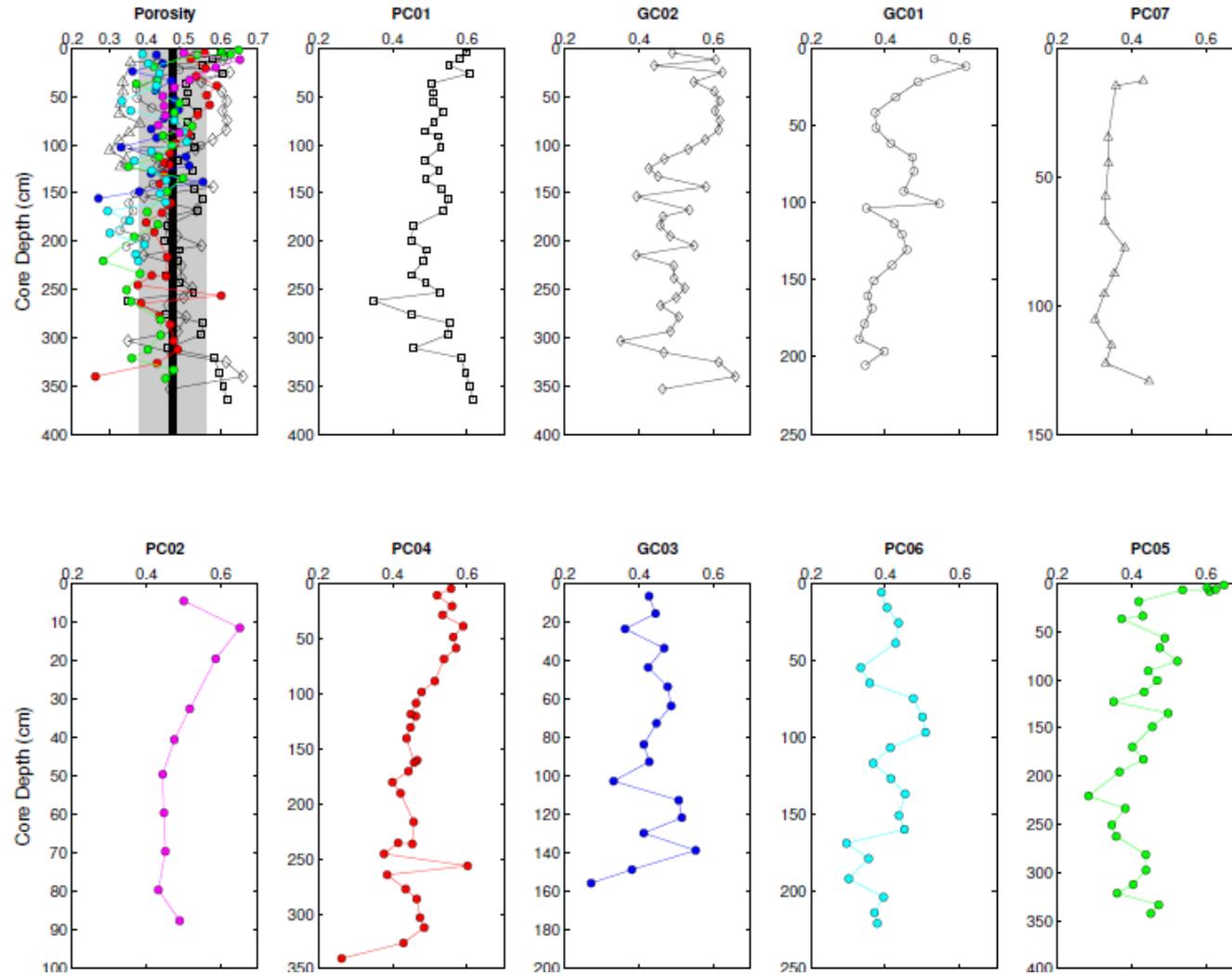
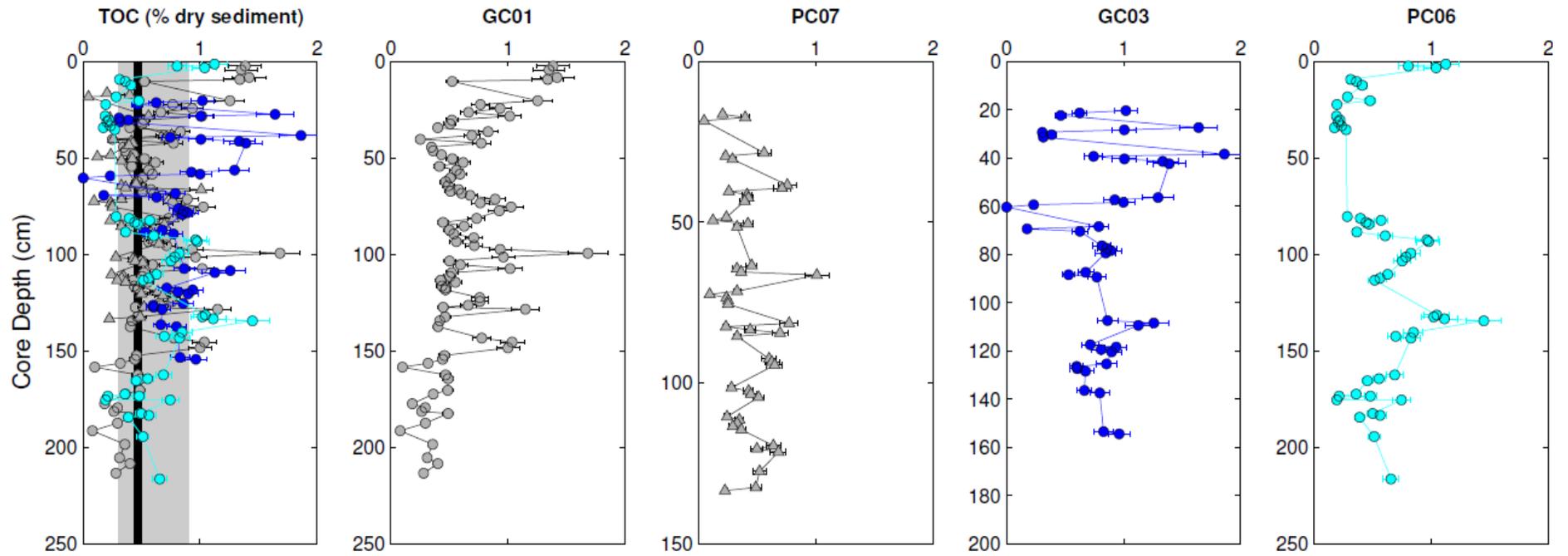


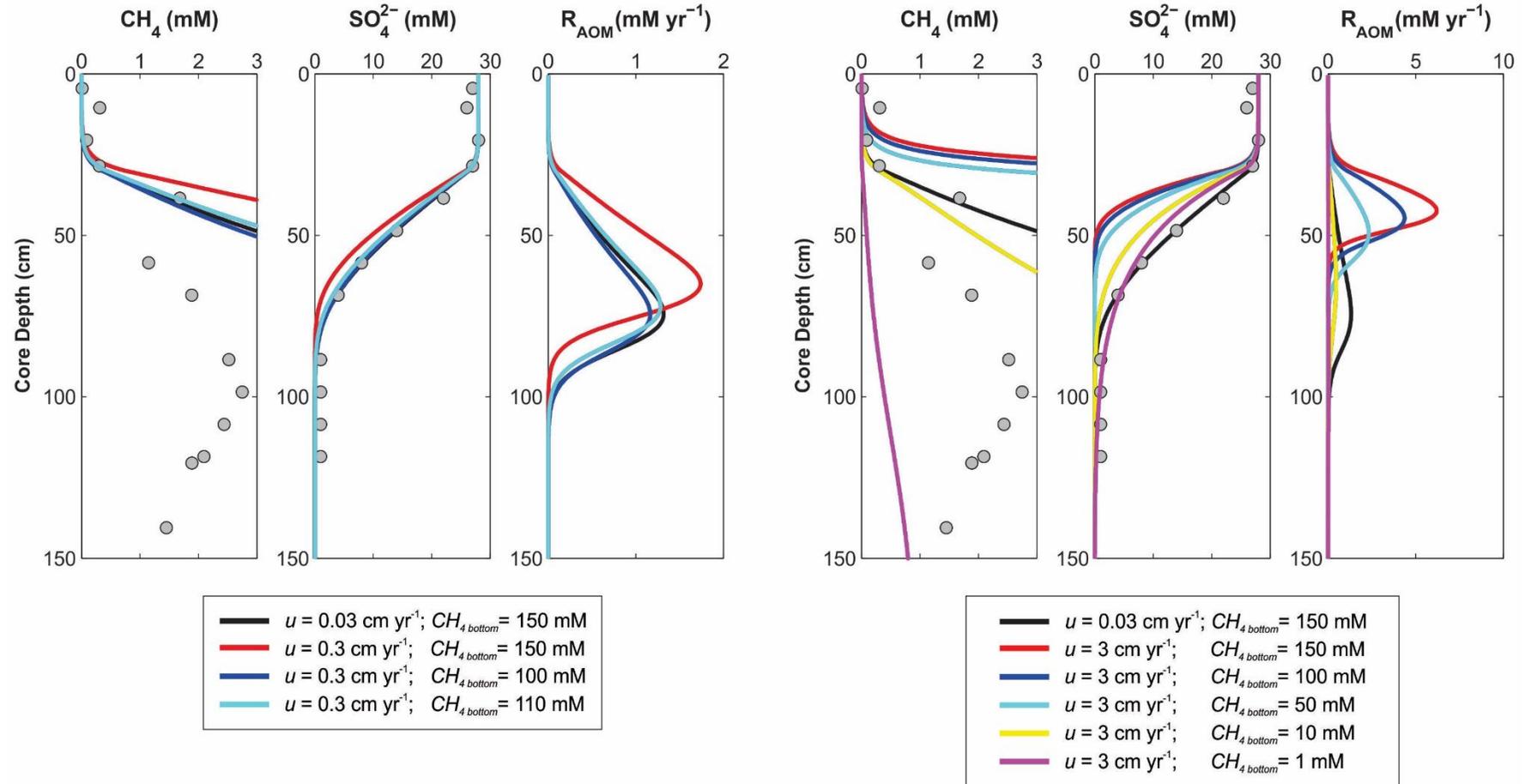
Supplemental Figure 1: Sediment core porosity profiles. In the upper left-hand panel the thick black vertical line indicates the average porosity (0.47) and the grey box the standard deviation (± 0.09).



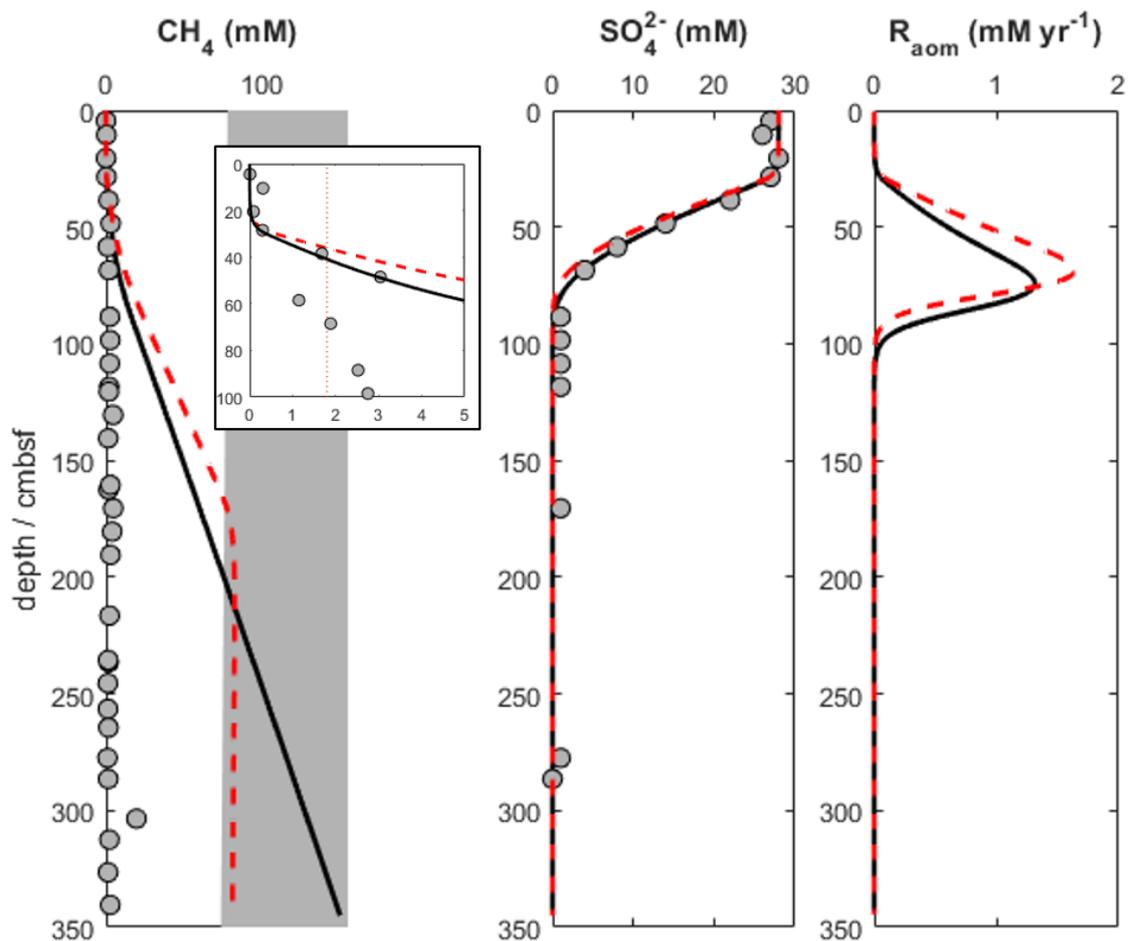
Supplemental Figure 2: Sediment core total organic carbon (TOC) profiles. In the left-hand panel the thick black vertical line indicates the average TOC (0.6 weight %) and the grey box the standard deviation (± 0.3 wt. %)



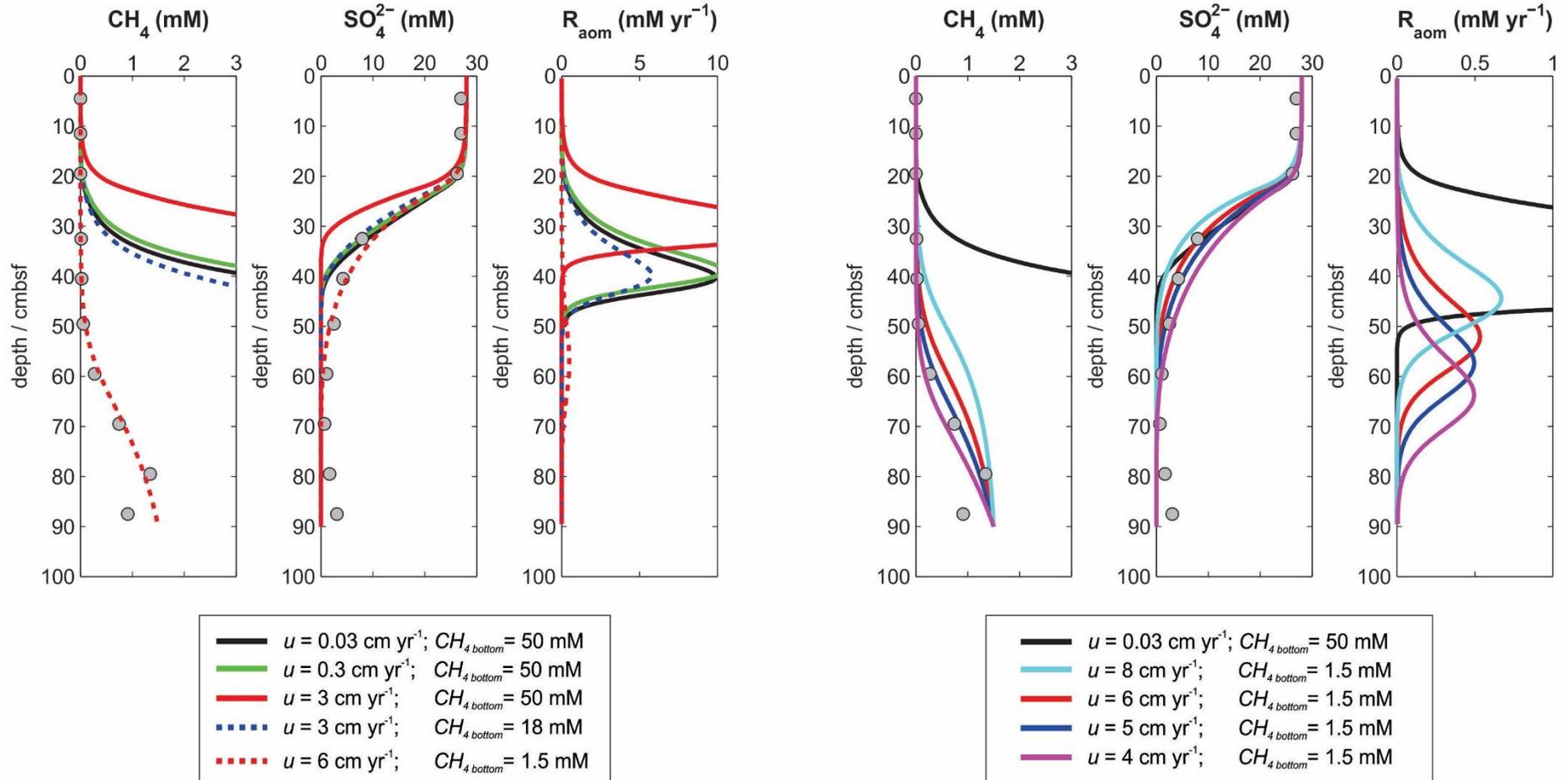
Supplemental Figure 3: Effect of advective flow rates (u) and core-bottom methane concentration (CH_4_{bottom}) on transport-reaction modelling results for core PC04. The black lines indicate the results presented in the main text (Fig. 5, Table 4). Corresponding model output methane fluxes and reaction rates are given in Supplemental Table 2. Grey circles indicated measured data (CH_4 and SO_4 only).



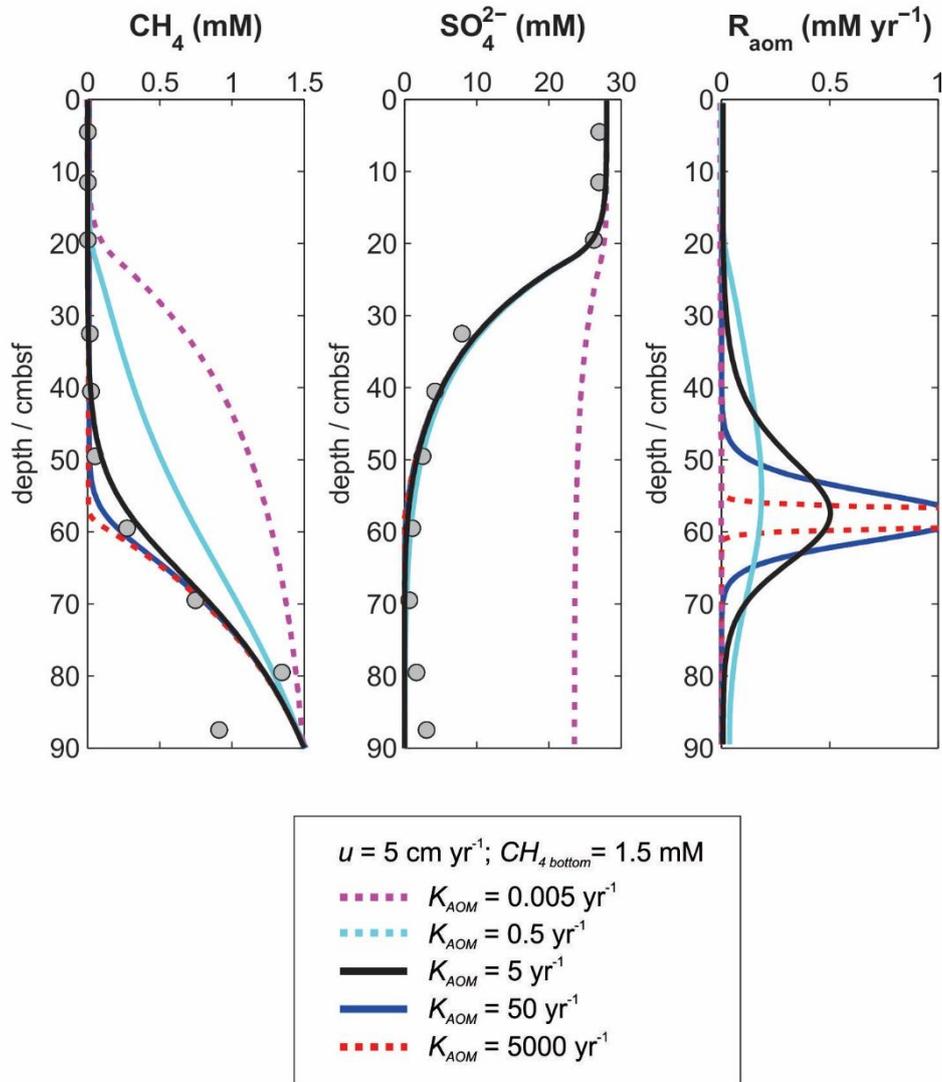
Supplemental Figure 4: Effect of including gas-phase methane transport at depths where modelled methane concentrations exceed saturation (~80 mM for this site) in core PC04. This was done by adding an upwards gas phase advection term (taken from Meister et al., 2013), to Equation 4: $A \cdot v_{gas} \cdot \delta(C(x) - C_{saturation(x)}) / \delta x$; where $C(x)$ is the methane concentration at depth x , $C_{saturation(x)}$ is the saturation concentration estimated following Dale et al. (2008a), v_{gas} is the gas rise velocity, and A is equal to 1 when $C(x) > C_{saturation(x)}$ and 0 when $C(x) < C_{saturation(x)}$. The solid black line is the best fit to the data without considering gas phase methane (Fig. 5, Table 4). Dashed red line shows the results of including gas phase transport with a v_{gas} of 10 cm/year (thin dash, conservative v_{gas} value used in Meister et al., 2013) and reducing the bottom boundary methane concentration from 150 mM to 81 mM. The grey shaded region indicates methane supersaturation, i.e. the presence of free gas. In the modified model the free gas is allowed to rise rapidly through the sediment column until it re-dissolves where methane is undersaturated. Setting the bottom boundary condition to a value higher than methane saturation means that methane gas is introduced from beneath the depth of the core. Corresponding model output methane fluxes and reaction rates are given in Supplemental Table 3. Grey circles indicate measured data (CH_4 and SO_4 only). The figure shows that adding the gas phase term and adjusting the imposed bottom boundary methane concentration has only a small effect on the rate of AOM, which is not surprising because AOM only acts on methane in the dissolved phase.



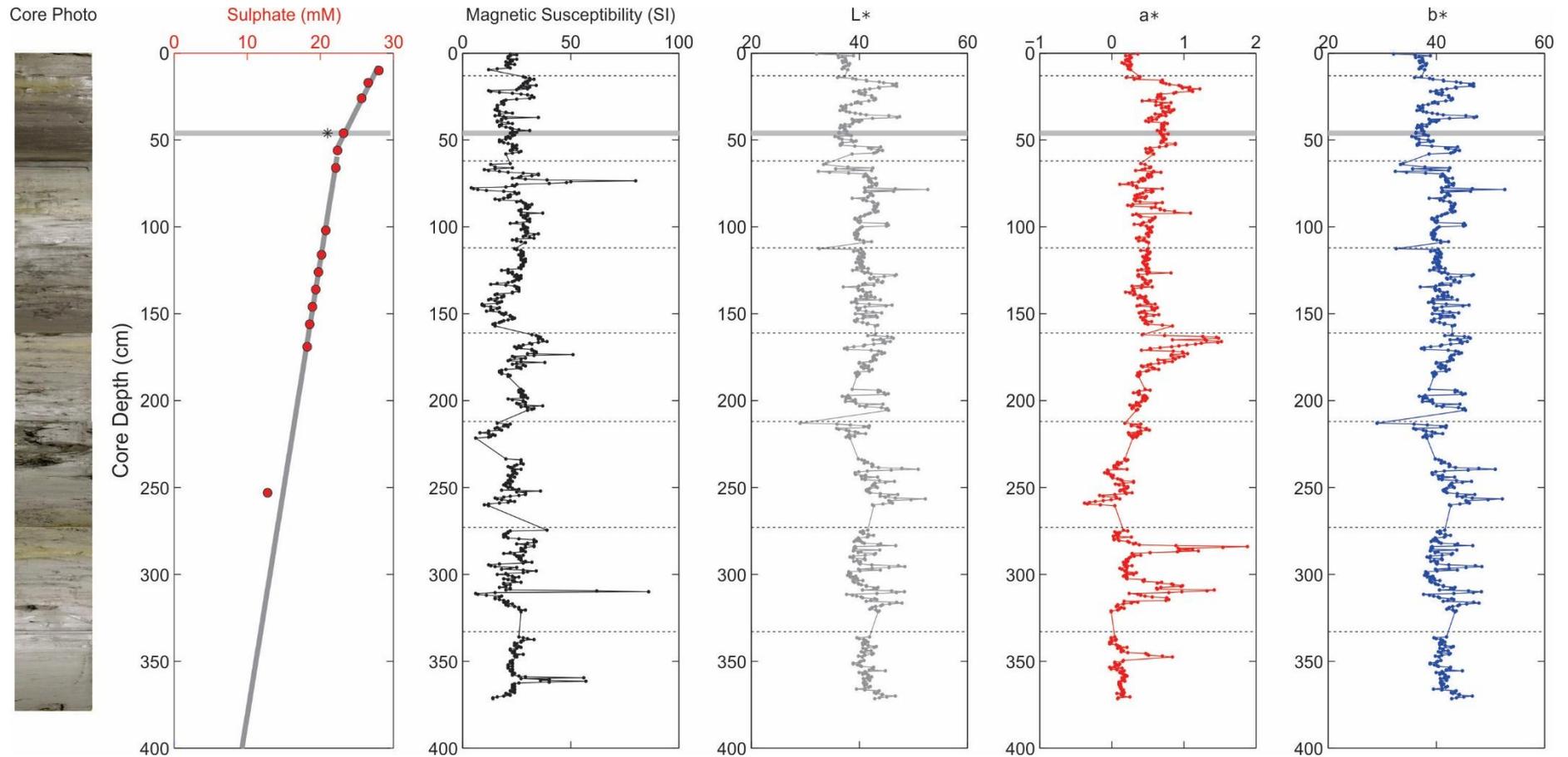
Supplemental Figure 5: Effect of advective fluid flow rates (u) and core-bottom methane concentration (CH_4_{bottom}) on transport-reaction modelling results for core PC02. The black lines and solid blue line indicate the results presented in the main text (Fig. 5, Table 4). Corresponding model output methane fluxes and reaction rates are given in Supplemental Table 3. Grey circles indicated measured data (CH_4 and SO_4 only).



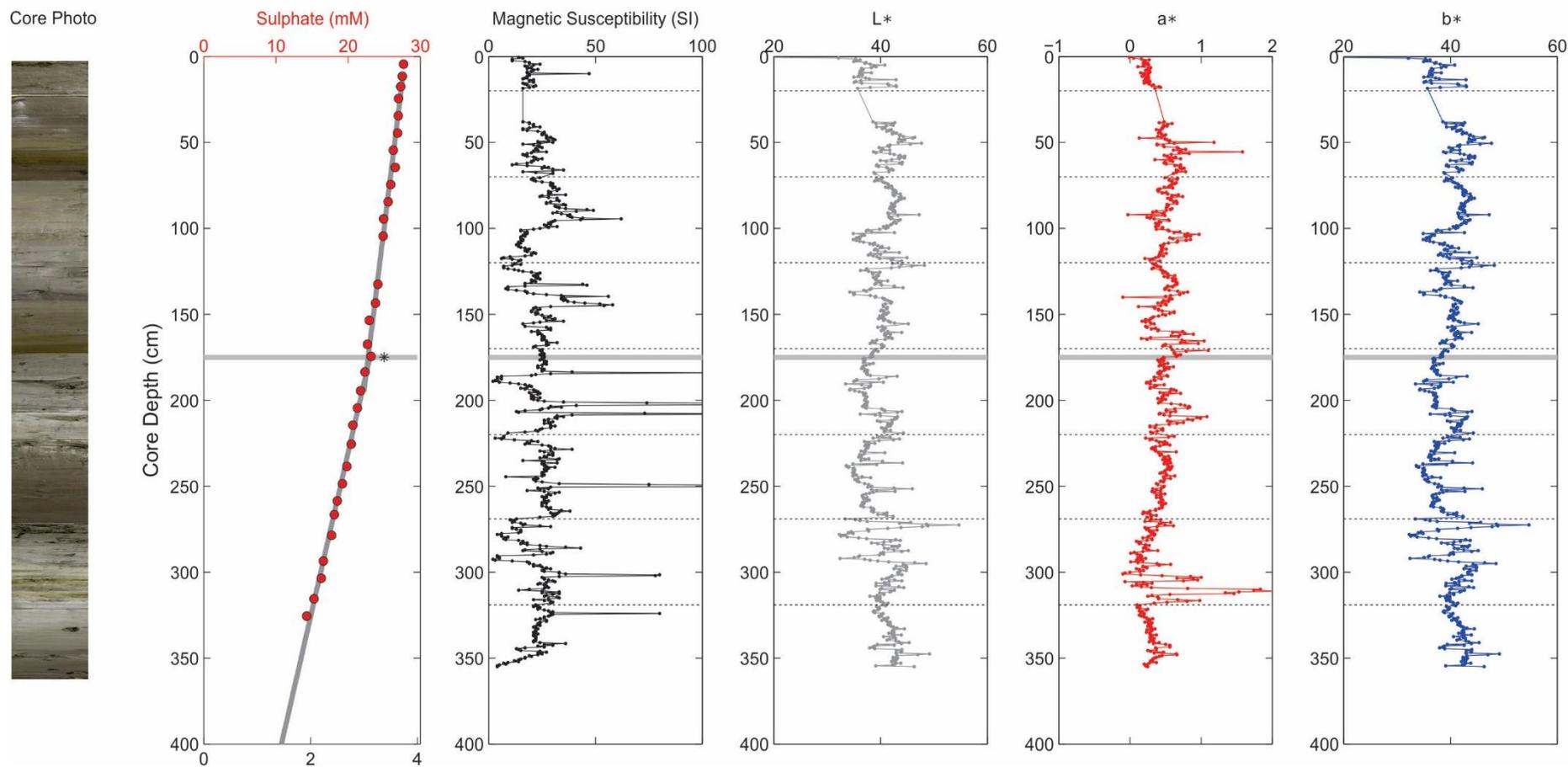
Supplemental Figure 6: Effect of the AOM rate constant (K_{AOM}) transport-reaction modelling results for core PCO₂ in conjunction with high advection (u) and low core-bottom methane concentration ($CH_{4\text{ bottom}}$). The black line indicate the results presented in the main text (Fig. 5, Table 4). Corresponding model output methane fluxes and reaction rates are given in Supplemental Table 3.



Supplemental Figure 7: Supplemental data for core PC01. From left to right: photo, pore water sulphate concentration profile showing position of “kink” in in linearity (star and solid horizontal line), Magnetic Susceptibility ($\times 10^{-5}$ SI)), and colour parameters L^* , a^* and b^* . Solid phase data determined on an XYZ Multi-Sensor Core Logger (Geotek) at 0.5 cm intervals. Dashed horizontal lines indicate divisions between core sections (~ 50 cm).



Supplemental Figure 8: Supplemental data for core GC02. From left to right: photo, pore water sulphate concentration profile showing position of “kink” in linearity (star and solid horizontal line), Magnetic Susceptibility ($\times 10^{-5}$ SI), and colour parameters L^* , a^* and b^* . Solid phase data determined on an XYZ Multi-Sensor Core Logger (Geotek) at 0.5 cm intervals. Dashed horizontal lines indicate divisions between core sections (~ 50 cm).



Supplemental Table 1:

Effects of porosity variation on transport-reaction modelling outputs.

parameter	units	GC03			PC02			
		0.5	0.4	0.6	0.5	0.6	0.4	piecewise*
porosity (f)	-	0.5	0.4	0.6	0.5	0.6	0.4	piecewise*
Output Parameters								
Methane Flux to Shallow Subsurface Sediments	mmol m ⁻² yr ⁻¹	120	160	80	550	750	390	500
Methanogenesis	mmol m ⁻² yr ⁻¹	0.21	0.17	0.25	0.17	0.14	0.20	0.17
AOM Rate	mmol m ⁻² yr ⁻¹	120	160	82	540	731	381	490
SO ₄ Reduction Rate for OM Remineralisation	mmol m ⁻² yr ⁻¹	0.30	0.25	0.35	0.13	0.11	0.15	0.13
Methane Flux to Seawater by Irrigation	mmol m ⁻² yr ⁻¹	0.005	0.010	0.003	10	17	5.4	4.9
Methane Flux to Seawater by Diffusion	mmol m ⁻² yr ⁻¹	0.0004	0.0005	0.0004	0.01	0.031	0.003	0.007

*piecewise porosity function for PC02, where x is sediment depth below seafloor in cm:

depth range (cm)		porosity equation	fit R ²	# data points fit
start	end			
0	5	0.5007	-	1
5	12	0.0216 x +0.4037	-	2
12	50	-0.0054 x +0.7017	0.98	5
50	70	0.0004 x +0.4254	1	3
70	80	-0.0019 x +0.5822	-	2
80	88	0.007 x -0.1357	-	2

Supplemental Table 2:

Effect of pore water advection and core bottom methane concentration on reaction transport model results for PC04. Highlighted columns indicate values which provided a good fit to measured methane and sulphate data (see supplemental figure 3).

parameter	units										
Pore Water Advection (u)	cm yr ⁻¹	0	0.03	0.3	3	0.3	0.3	3	3	3	3
Core Bottom Methane Concentration ($C_{CH4\ bottom}$)	mM	150	150	150	150	100	110	100	50	10	1
<i>Output Parameters</i>											
Methane Flux to Shallow Subsurface Sediments	mmol m ⁻² yr ⁻¹	301	310	417	2251	284	311	1500	750	150	15
Methanogenesis	mmol m ⁻² yr ⁻¹	0.89	0.89	0.92	1.00	0.89	0.90	0.99	0.97	0.91	0.83
AOM Rate	mmol m ⁻² yr ⁻¹	239	240	282	523	221	234	422	284	95	13
SO ₄ Reduction Rate for OM Remineralisation	mmol m ⁻² yr ⁻¹	0.25	0.25	0.22	0.15	0.25	0.24	0.15	0.17	0.23	0.31
Methane Flux to Seawater by Irrigation	mmol m ⁻² yr ⁻¹	63	70	137	1728	64	77	1079	467	56	3
Methane Flux to Seawater by Diffusion	mmol m ⁻² yr ⁻¹	0.02	0.02	0.04	0.6	0.02	0.02	0.4	0.2	0.0	0.0

Supplemental Table 3:

Effect of including a gas-phase transport term for PCO₄ (see also supplemental figure 4). When the gas phase transport term is included, setting the bottom boundary methane concentration above saturation (~80 mM), implies adding a gas-phase methane input into the base of the modelled sediments. Note that the effects of including the best fit gas-phase transport term on rates of AOM and methane fluxes across the sediment-seawater interface are small.

parameter	units		
Core Bottom Methane Concentration ($C_{CH4\ bottom}$)	mM	150	81
Gas Rise Velocity ($v_{C_{gas}}$)	cm yr ⁻¹	0	10
<i>Output Parameters</i>			
Dissolved Methane Flux to Shallow Subsurface Sediments	mmol m ⁻² yr ⁻¹	310	-5
Gas Phase Methane Flux to Shallow Subsurface Sediments	mmol m ⁻² yr ⁻¹	-	380
Methanogenesis	mmol m ⁻² yr ⁻¹	0.89	0.91
AOM Rate	mmol m ⁻² yr ⁻¹	240	280
SO ₄ Reduction Rate for OM Remineralisation	mmol m ⁻² yr ⁻¹	0.25	0.23
Methane Flux to Seawater by Irrigation	mmol m ⁻² yr ⁻¹	70	110
Methane Flux to Seawater by Diffusion	mmol m ⁻² yr ⁻¹	0.02	0

Supplemental Table 4:

Effect of pore water advection, core bottom methane concentration, and AOM rate constant on reaction-transport model results for PCO2. Highlighted columns indicate values which provided the best fits to measured methane and sulphate data (see supplemental figures 5 and 6).

parameter	units												
Pore Water Advection (u)	cm yr ⁻¹	0.03	0.3	3	3	6	8	5	4	5	5	5	5
Core Bottom Methane Concentration ($C_{CH4\ bottom}$)	mM	50	50	50	18	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
AOM Rate Constant (K_{AOM})	yr ⁻¹	5	5	5	5	5	5	5	5	0.005	0.5	50	5000
<i>Output Parameters</i>													
Methane Flux to Shallow Subsurface Sediments	mmol m ⁻² yr ⁻¹	550	570	937	362	52	63	50	50	40	48	50	50
Methanogenesis	mmol m ⁻² yr ⁻¹	0.2	0.2	0.2	0.2	0.1	0.2	0.1	0.1	0.0	0.1	0.1	0.1
AOM Rate	mmol m ⁻² yr ⁻¹	540	557	820	353	52	62	50	50	2	38	50	50
SO ₄ Reduction Rate for OM Remineralisation	mmol m ⁻² yr ⁻¹	0.1	0.1	0.1	0.1	0.2	0.1	0.2	0.2	0.3	0.2	0.2	0.2
Methane Flux to Seawater by Irrigation	mmol m ⁻² yr ⁻¹	10	13	117	9	0.2	1	0.09	0.03	38	9.8	0.0004	5 × 10 ⁻⁶
Methane Flux to Seawater by Diffusion	mmol m ⁻² yr ⁻¹	0.01	0.01	0.15	0.01	0.001	0.002	0.0004	0.000	0.16	0.04	0.0001	5 × 10 ⁻⁶