- Long-Term Atmospheric Emissions for the Coal Oil Point Natural Marine
 Hydrocarbon Seep Field, Offshore California
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Supplemental Material

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9 S1 The Gaussian Plume model theoretical basis

Sea surface emissions for focused bubble plumes can be described as a point source whose atmospheric plume can be modeled as a Gaussian plume (Hanna et al., 1982). The Gaussian plume model relates atmospheric emissions, E_A , to the concentration anomaly, C', relative to ambient, C, wind speed, u, and the atmospheric turbulence parameters, σ_y and σ_z , defined in a cartesian coordinate system where x is the downwind, i.e., wind direction, θ , y is the transverse direction, and z is the vertical coordinate,

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$$C'(x, y, z) = E_A \frac{exp\left(\frac{-y^2}{2\sigma_y^2}\right)}{(2\pi u \sigma_z \sigma_y)} \left(exp\left(\frac{-(z-h)^2}{2\sigma_z^2}\right) + exp\left(\frac{-(z+h)^2}{2\sigma_z^2}\right) + exp\left(\frac{-(z-2(BL-h))^2}{2\sigma_z^2}\right) + exp\left(\frac{-(z-2(BL-h))^2}{2\sigma_z^2}\right$$

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$$exp\left(\frac{-(z+2(BL-h))^2}{2\sigma_z^2}\right)$$
, (S1)

18 where *h* is the measurement height. The second exponential term represents reflection off the sea 19 surface and assumes a non-sticky molecule, the third term represents reflection off the marine 20 boundary layer, at height *BL*, and the fourth term represents re-reflection off the surface (**Fig. S1**). A 21 shallower boundary layer height, *BL*, contributes more significantly to this reflection, for example, a 22 250-m *BL* and u = 2.1 m s⁻¹ contributes 10-20% of *C*' from 2-km downwind (**Fig. S1**). *C*' is relative 23 to the background concentration, *C*, which is based on concentrations outside the plume's edges and 24 can include environmental gradients from large-scale transport processes. 25



Figure S1: Surface concentration, C', for Gaussian plumes for $u=2.1 \text{ m s}^{-1} \text{ A}$) with no planetary boundary layer, BL, B) reflection from a 250-m BL (Eqn. 1, terms 3+4) and C) Gaussian plume with 250-m BL. D) Reflection plume from a 100-m BL and E) Gaussian plume with 100-m BL. Arbitrary C' units. Data key on panel.

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31 Parameterization of σ_y and σ_z are:

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$$\sigma_v = a(1+10^{-4}x)^{-1/2}; \sigma_z = b(1+cx)^n$$
, (S2)

33 with a, b, and c depending on stability class (Briggs, 1973). Stability class (Table S2) is based on solar insolation, I, and u (Hanna et al., 1982). These turbulence parameterizations (Eqn. S2) are 34 35 discrete (Table S1), which introduces uncertainty in σ_x and σ_y , particularly at stability class transitions. To remove discretization distortions, a 2nd-order polynomial was fit to the turbulence 36 37 parameters a, b, and c, for each insolation class with respect to u (Fig. S2). The polynomial fit then 38 was evaluated for u to determine a, b, and c for all three insolation classes. Strong, moderate, and weak insolation were fit for class values of I = 800, 500, and 175 W m⁻², respectively. The 39 parameterizations are shown in Fig. S2 and are for $1 \le u \le 6$ m s⁻¹. Given the coarse nature of the 40 41 Pasquill classes, no effort was made to optimize; simply, the center u for each class was used in the 42 parameterization.

C1		unneter	, 101 au	iospii o n		
Class		a	b	С	n	Class Description
А	open	0.22	0.20	0	-	Extremely unstable
В	open	0.16	0.12	0	-	Moderately unstable
С	open	0.11	0.08	2x10 ⁻⁴	-0.5	Slightly unstable
D	open	0.08	0.06	0.015	-0.5	Neutral
E	open	0.06	0.03	3x10 ⁻⁴	-1	Slightly stable
F	open	0.04	0.016	3x10 ⁻⁴	-1	Moderately stable
*Briggs (1973).						

44 Table S1. Parameters for atmospheric turbulence parameterizations for different stability classes*

54 **Table S2.** Conditions defining Pasquill turbulence classes*

55	и	Istrong	I _{Moderate}	IWeak	Nighttime	Nighttime
56	<u>m s⁻¹</u>	>700 W m ⁻²	350-700 W m ⁻²	<350 W m ⁻²	> ¹ / ₂ clouds	< ³ / ₈ cloudiness
57	<2	А	A-B	В	E	F
58	2	A-B	В	С	E	F
59	4	В	B-C	С	D	Е
60	6	С	C-D	D	D	D
61	>6	С	D	D	D	D

62 *from Hanna et al. (1982). *u* is wind speed. *I* is solar insolation.

63

64

The Gaussian plume model is for a passive dispersant and assumes that negligible along-wind diffusion, that u, σ_y , and σ_z are vertically and horizontally uniform along its trajectory, that ufluctuations are zero, and that u remains parallel to the x axis (no veering). These assumptions imply an idealized, flat terrain of homogeneous roughness. Violations of these conditions are common in terrestrial settings and can be addressed by model modification, e.g., Briggs (1973); Leifer et al. (2016), the absence of topography and obstacles at sea implies that marine plumes generally satisfy the Gaussian plume requirements.



73 **Downwind distance (m)** 74 **Figure S2:** Pasquill turbulence parameters (**Table S1**) and 2nd-order polynomial least-squares linear-regression fits of **A**) 75 a, **B**) b, and **C**) c for solar insolation stability classes (**Table S1**; class data key on figure) versus wind speed, u. See text 76 for details. **D**) Gaussian plume surface concentration, C', for 0.47 L min⁻¹ emissions, u = 3.5 m s⁻¹, and no boundary layer.



84 S2 Focused seep areas

86	<u>Seep Area</u>	heta	Latitude, Longitude
87	La Goleta Seep	152°	34° 23.503'N, 119° 51.193'W
88	Seep Tent Seep	198°	34° 23.063'N, 119° 53.428'W
89	Platform Holly	238°	34° 23.392'N, 119° 54.374' W
90	Trilogy Seep C	178°	34° 23.634'N, 119° 52.702'W
91	Trilogy Seep B	178°	34° 23.620'N, 119° 52.709'W
92	Trilogy Seep A	178°	34° 23.603'N, 119° 52.699'W
93	Patch Seep	140°	34° 21.850'N, 119° 49.755'W
94	Shane Seep	230°	34° 24.370'N, 119° 53.428'W
95	IV Super Seep	146°	34° 24.090'N, 119° 52.066'W
96	Tonya Seep	184°	34° 24.043'N, 119° 52.841'W
97	Horseshoe Seep	186°	34° 23.799'N, 119° 52.519'W
98	Rostocker Seep	99°	34° 24.230'N, 119° 50.438'W
99	Seadog Seep	240°	34° 24.172'N, 119° 54.374'W

85 **Table S3.** Location and direction from West Campus Station of informally-named seeps

100 * θ - direction from West Campus Station (34.414949°N, 119.879690°W).

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102 S3 Sonar return

103 The Sept. 2005 seep field survey sonar return, ω , was gridded at spatial resolutions from 11 to 104 225 m in a coordinate system with origin at West Campus Station, WCS, 34° 24.8969'N, 119° 105 52.7814'W (**Fig. S3**). See Leifer et al. (2010) for details on the sonar survey data acquisition and 106 analysis. Gridding involves averaging of all ω in each bin followed by a gap-filling low-pass filter. 107 To gap fill, the center bin in a rolling 3x3-bin window which is empty, is replaced by the mean if 108 there are more than 5 non-empty bins in the window. A hybrid 56/22-m gridding scheme gap filled 109 the 22-m grid with the 56-m grid and was used in simulations (**Fig. S3C**).



 $\begin{array}{c} 111\\ 112 \end{array}$ Figure S3: Sonar return, ω, maps for gridding at A) 11-m, B) 22 m, C) 22/56-m D), 56 m, E) 110 m, and F) 225 m. See 113 text for details. Data key on panels.

115 The noise level was 0.015 and was identified from a histogram of ω , where the ω probability 116 distribution shows a power law shift and is the transition from seep distribution to noise domination 117 (Fig. S4). Although there clearly is a seep contribution below the noise level, it cannot be segregated 118 from noise and is neglected.

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Figure S4: A) 22/56-m gridded 2005 sonar return, ω . Green dot shows West Campus Station (WCS) location. Shown in the \mathbb{C} Google Earth environment. B) Occurrence distribution, $P(\omega)$. Arrow shows noise level based on shift in power law 123 fits to P at ω~0.015. Data key on panels. SBA - Santa Barbara Airport.

125 S4 Gaussian Plume model of an area plume

The model's core routines calculate a Gaussian plume (**Fig. S5A**) for the specified meteorological conditions in a Cartesian coordinate system. Then, the plume is rotated to the wind direction, regridded, and normalized to ensure mass conservation (**Fig. S5B**). The rotated plume is translated to each grid cell, scaled to emissions for that grid cell, and finally, all plumes are added (**Fig. S5C**). *C*' at WCS is calculated from the combined seep field emission plume map (**Fig. S5D**).

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132Easting (km)Easting (km)133Figure S5: A) Gaussian plume concentration (arbitrary units) for 4 m s⁻¹ wind speed and 250 m-thick boundary layer. B)134Plume rotated to a wind direction, θ , of 135°, regridded, and normalized. C) Assembled plumes from all grid cells. Dashed135red line shows a constant northing transect through West Campus Station, WCS. D) Concentration profile along WCS136transect (dashed red line in panel C). Red line shows location of WCS.

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138 S5 Field atmospheric observations

Winds for 28 May 2016 were largely from the east with some modifications near the coast (**Fig. S6**). Winds in Goleta Bay included a strong onshore component, which was absent offshore Isla Vista where the shoreline features tall bluffs (~10-14 m). To the west of COP, winds had a strong offshore component as far as halfway to Naples. Enhancements in the transect offshore west Sands Beach and Haskell Beach are consistent with emissions from shallow waters, beaches, and/or coastal onshore from seepage and/or abandoned oil wells.



146 C_{CH4} (ppm) Storm u (m s⁻¹) UCSB – Univ Calif Santa Barbara
 147 Figure S6: Methane, C_{CH4}, and winds, u, for 28 May 2016. Sonar return map for reference in background. Dashed arrow shows proposed onshore winds; black arrows show travel direction. Shown in Google Earth environment. [©]Google Earth.
 149 See Table S3 for locations of informally named seeps.

On 28 May 2016 winds were strongly from the east, which is uncommon, and largely overwhelmed the sea breeze. The easterly winds flow onshore in Goleta Bay and then offshore around Ellwood and Sands Beach, to the west of COP. This likely relates to a combination of the sea breeze in Goleta Bay and the overall easterly flow which in turn drives an offshore flow (by continuity) around Ellwood and Haskell Beaches. This wind flow creates strong convergence and divergence air flows.

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158 S6 West Campus Station data

159 **S6.1 Trends**

WCS data show a significant change in 2008 when the station was upgraded from 1-hour to 1minute time resolution, allowing far higher values of total hydrocarbon concentration, THC, and u(Fig. S7). Hereafter, all concentrations and emissions are THC unless otherwise noted. To assess whether the station upgrade introduced biases, $\phi(u)$ prior and post 2008 were compared (Fig. S7C, 164 **7D**). The slopes of $\phi(u)$ with respect to *u* for 2 to 4 m s⁻¹ are nearly identical for ϕ for the pre- and 165 post-2008 data, the main change is for highest and weakest *u*, i.e., the range was extended. Moreover, 166 several shifts between subsequent years for *u* and *C* have been documented (1997 and 2012 for *u*, 167 1997 and 2013 for *C*), i.e., such shifts in annualized values are not unique to 2008 (**Fig. S8**). This 168 strongly argues that the changes in average, median, and baseline values post 2008 were from better 169 measurement of gusts and weak winds and short positive *C* anomalies.





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A linear regression analysis of the wind data showed increasing median u of 0.023 m s⁻¹ yr⁻¹ and 0.04 m s⁻¹ yr⁻¹ for all directions and for seep directions, respectively (**Fig. S9A**). This corresponds to an increasing sea breeze (from the seep direction). Summer winds have increased faster than winter winds, 0.043 versus 0.027 m s⁻¹, respectively.

The residual, *R*, to the Gaussian fit to $C'(\theta)$ showed an overall increasing linear spatial trend of 0.17 ppb degree⁻¹ from 175° to 210° (**Fig. S9B**). There is a peak at *R*~230° that corresponds to Shane Seep and a peak at *R*~170°, which does not correspond to any named seeps (**Table S3**), but is in the general direction of the inshore seep trend to the east of the Trilogy Seeps (**Fig. 1**).



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Figure S9: A) Seasonally-segregated, annually-averaged wind speed, *u* for WCS. Summer is for Julian day 135-250, winter is Julian day 260-130, and three-year rolling-average. **B)** Residual, *R*, to dual Gaussian plume fit (**Fig. 4A**) to winddirection resolved concentration, $C(\theta)$, for 1990-2019 for unsmoothed, 5° smoothed, and least-squares, linear-regression analysis fit. Data key on panel.

195 **S6.2 Diurnal**

Hourly-segregated averaged u and C for 1991-2007 (Fig. S10) show similar overall patterns to 2008-2021 for the seep directions (Fig. 6). The station improvements shifted averages, making averaging the entire datasets inappropriate. One difference was the late-afternoon peak in C towards the east-northeast, which was absent in the earlier period. The significant expansion of housing and commercial developments in this direction in recent decades likely underlies this change.

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202 θ (°) Hour 203 Figure S10: 1991-2007 averaged wind direction, θ , and hourly-resolved A) wind speed, u, and B) concentration, C. C) 204 Seep-direction (90–270°), hourly-averaged wind speed, u_{seep} , and D) concentration, C_{seep} , averaged, individual years, and 205 3-year smoothed. Data key on figure. Midnight data missing due to daily calibration.

206

207 S7 Seep field emissions

208 S7.1 Effect of model wind direction resolution

Emissions simulations are run iteratively, initialized by the sonar distribution, which was multiplied by a scaling factor, $K(\theta)$, initialized with K=22,620, that was applied to E(x,y). Notably, the θ -resolved C'_{sim} and C'_{obs} disagree significantly. This is unsurprising as the relationship between ω and seabed emissions is complex (Leifer et al., 2017) with additional complexity arising from transport across the water column. For sensitivity studies, the scaling factor, $K(\theta)$, see Eqn. 3, was applied to $E(\theta)$ after each iteration until convergence (<1%), typically within 5 iterations (Fig. S11).



Figure S11: Observed and simulated WCS concentration anomaly, C'_{obs} and C'_{sim} , respectively, versus wind direction, θ , for all iterations for 22/56-m sonar map and angular, θ , resolutions, of **A**) 10°, **B**) 5°, **C**) 2°, and **D**) 1°. Data key on figure.

Simulations were conducted with no distance correction, i.e., $K(r, \theta) = 1$ for north offsets of WCS location, *Y*, from 0 to 3 km. For reference, the inner and outer seep trends are 1.5 and 3.5 km from WCS at their closest direction. Simulations showed a near linear response in E_A to *Y* (**Fig. S12**), which was applied to as a distance scaling function, $K(r, \theta)$, in all further simulations.



Figure S12: Simulated atmospheric emissions, E_A , with respect to north offset, Y, modeled with no distance varying correction factor, and linear fit shown on panel.



 ω (-) 229 Figure S13: Sonar return, ω , versus derived atmospheric emissions, E_A . Line illustrates unity power law.

231 S7.3 Sonar return versus atmospheric emissions

232 Plotting all E_A versus ω shows a non-linear relationship (Fig. S13) with E_A increasing very steeply with ω before rolling over for values of E_A in the range 1-10 g s⁻¹ m⁻². After rolling over, there were 233 234 several populations of ω -values that increased as a power law with an exponent slightly less than 235 unity. Emissions are the fraction of the seabed emissions that are transferred into the atmosphere, 236 including from adjacent pixels where emissions are from evasion - the simulation was initialized with 237 non-zero ω in neighboring cells to allow the simulation to infer sear-air gas evasion emissions from these cells. This accounts for values of ω less than 0.02 in Fig. S13. The highest ω corresponds to the 238 239 strongest plumes where sonar tends to saturate (Leifer et al., 2017) but dissolved gas evasion 240 emissions to the atmosphere are more efficient due to the upwelling flow, which is more efficient in 241 strong plumes.

242

243 S7.4 Emissions Uncertainty

244 S7.4.1 Angular resolution

Sensitivity to angular resolution, $\delta\theta$, is very weak, <1% for reducing $\Delta\theta$ from 10° to 1°. For example, a resolution of $\delta\theta$ =1° produced artifacts and only changed *E* by 0.3% for reducing $\delta\theta$ from 2° to 1°. $\delta\theta$ resolution affects simulation resolution with coarse resolution blurring the fine-scale sonar structure that is apparent in the 1° simulations (**Fig. S11D**); however, 1° simulations produced overly-quantized results for the hybrid 22/56-m sonar maps including banding artifacts. Thus, θ resolution was 2° outside of several sensitivity simulations.

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252 S7.4.2 Veering

Veering, ψ , was implemented by adding ψ to θ in the calculation of $K(r, \theta)$, i.e., $K(r, \theta+\psi)$. Wind veering emphasized emissions at the field's edges, clearly affecting the seep emissions map. $\psi=+10^{\circ}$ produced better agreement with the sonar map. Wind veering simulations (**Fig. S14**) overly emphasized emissions at the seep field's edge towards which there was veering, although the no-wind veering simulation also emphasized the western field's seepage. Overall, the $\psi =+10^{\circ}$ was an improvement. Specifically, it improved the importance of La Goleta Seep compared to the no wind veering simulation and also lessened the enhancement of the western seep field's edge (**Fig. 7B**).



260 261 262 Figure S14: A) Sonar return, ω , map gridded to 56-m resolution. B) Atmospheric emissions, E₄, map for wind veering, ψ , of **B**) ψ =-10° and **C**) ψ =10°. Data key on figure.

E_A was extremely weakly sensitive to ψ , with $\pm 10^{\circ}$ veering corresponding to only a few percent in *E_A*. This low sensitivity arises because ψ simply shifts emissions to neighboring cells, only changing *E_A* when neighboring cells have no seepage forcing the model to increase emissions from more distant cells.

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268 **S7.4.3 Wind speed**

269 There is a strong sensitivity of E_A to u, particularly for small negative variations, although 270 sensitivity saturated at $\pm 10\%$ with greater sensitivity for lower *u*. This sensitivity was shown by a 271 series of non-iterative simulations of $C'(\theta)$ for a range of u, showing variations were most strong for very weak wind speeds. Observed u span the range of weak for the east and south to moderate for 272 273 prevailing (Fig. S15A). Instrumentation uncertainty in *u* is a few percent, thus uncertainty in *u* arises 274 if WCS winds are unrepresentative due to acceleration or deceleration between the seep field and WCS. C' is highest for $u = 1 \text{ m s}^{-1}$, which is weaker than actual winds, whereas C' for $u = 8 \text{ m s}^{-1}$ is 275 about a tenth C' for $u = 1 \text{ m s}^{-1}$. At lower u, there is more "blurring" of plumes from different sources 276 277 showing less defined structure than higher u. However, u and BL are not independent – by continuity, 278 an increase in u – i.e., acceleration, requires a decrease in *BL*. Thus, *BL* sensitivity largely counters u279 sensitivity, discussed in Sec. S8.5.

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285 S7.4.4 Boundary layer

286 BL sensitivity is weakly non-linear and not strong - a 40% reduction in BL (250 to 150 m) only 287 decreased E_A by 17%, less so for a 100 m increase to 350 m in *BL* (Fig. S15B). The effect of large BL changes saturates – for very high BL, it has no effect, and for very shallow BL, multiple reflections 288 289 homogenize the boundary layer leaving only dilution driving BL sensitivity. Thus, a factor of 40 290 variation in *BL* corresponds to a 200% change in $C'(\theta)$. Although the nocturnal *BL* is significantly 291 shallower, nocturnal winds largely are the (northerly) land breeze and thus uncommon for the seep 292 direction. BL vales for the seep field and WCS are uncharacterized, thus, a 2nd-order polynomial was fit to *BL* versus E_A and averaged (absolute) over possible ranges. For 200 \leq BL \leq 300 m, the mean 293 294 absolute value was 3%, 6% for 150<BL<350 m.

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296 S7.4.5 Concentration

297 E_A sensitivity to C' is weaker than that of u, particularly for small variations and is linear (Fig. 9). 298 C uncertainty arises from drift in the flame ion detector between nightly calibrations. Thus, the 299 difference in concentration before and after calibration is the daily drift as well as any changes in 300 ambient, which should on average be zero, particularly for around midnight when the atmospheric is 301 stable. A 500-point histogram of the total daily drift was calculated and values $\leq \pm 0.2$ ppb were fit with a Gaussian function (larger deviations exhibited a broader distribution which, likely resulted 302 303 from ambient trends) and found a half-width of 170 ppb, or ± 85 ppb. Thus, the average drift is ~ 40 ppb dy⁻¹, reducing for annualized values to ~2 ppb dy⁻¹ ($1/\sqrt{365}$), i.e., <1% of the C' value of 300 304 305 ppb. As such, uncertainty in C' is not a significant contributor to uncertainty in E_A .

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307 S7.4.6 Combined wind and boundary Layer

 E_A sensitivity to *BL* largely counters E_A sensitivity to *u* (**Fig. 9D**), even when mismatched – a $\pm 40\%$ decrease in *BL* and $\pm 20\%$ increase in *u* corresponds to a 6% and 9% decrease in E_A , i.e., less than half the mismatch. By continuity, this should be quite small, reflecting lesser changes in pressure, temperature, and topographic forcing. Underlying this weak sensitivity is that the linear sensitivity of E_A to *u* largely arises from dilution as for *BL*. Note, this weak sensitivity is for a change in *BL* from the offshore to onshore and the resultant change on winds, not a change in the overall (regional) *BL*.



315 316 317 Figure S16: A) Sonar return, ω , map gridded to 56/22-m resolution. Atmospheric emissions, E_A, for inshore/offshore trends relative enhancement, ζ , for **B**) ζ =-25% and **C**) ζ =+25% offshore enhancement.

319 S7.4.7 Inshore / offshore partitioning

320 The model cannot determine how to apportion emissions between the inshore and offshore seeps. 321 Simulations found a complex relationship between E_A and ζ (Fig. 9) because simulations re-assign 322 some emissions between the seep trends depending on the presence or absence of seepage for θ where 323 there is no seepage on one of the two trends (Fig. S16). As such, a potentially strong sensitivity arises 324 from the offshore and onshore seepage trend partitioning with respect to θ . Initial simulations that 325 used a uniform distance partitioning, unrealistically increased inshore seepage compared to ω . Given 326 the similarity in the ω probability distribution between the inshore and offshore trends (Fig. 8), i.e., 327 whatever the functional relationship between E_A and ω is, similar ω implies similar E_A .

Sensitivity simulations were run for various shifts in the relative emissions from the inshore and offshore trends – i.e., for +10% inshore -10% offshore, while maintaining a linear variation in $K(r, \theta)$. Sensitivity is strong, and complex – interestingly, reversing between -10° and 10°. This behavior is due to shifting emissions onto different seep areas in the inshore or offshore seep area. As such, for - $30\% < \zeta <+25\%$, uncertainty ranges from -10% to +5%. Although higher changes in ζ are certainly feasible, both trends source from the same formation, which means on a 30-year time scale (or much shorter), changes in one seep trend likely reflect trends in all seep areas.

Additionally, seepage could shift within the trends – nearer or further - which was assessed by the simulations of north shifts of WCS (**Fig. S12**). Total trend widths are ~300 m for the offshore and 600 m for the inshore, or an average distance deviation of \pm 75 and 150 m, corresponding to E_A variations of a few percent.

339

340 S8 Future data needs and model improvements

341 S8.1 Key data needs

The driver of uncertainty was whether emissions arose from the inshore or offshore seep trends, which could not be determined from WCS data. A second air quality station located towards the seep field east side would allow triangulation. If the station included a ceilometer, it also could address *BL* uncertainty in E_A . Long-term *BL* data would characterize the relationship to solar insolation, diurnal cycle, and winds, allowing application to historical data. Additionally, THC is a less than ideal seep emission indicator as it can arise from other sources,
such as motor vehicles or fires – the station should include speciation, ideally a combination of CEAS
analyzers and a gas chromatograph.

The diurnal cycle is poorly constrained by WCS data given that the typical diurnal cycle of strengthening winds in the afternoon also shifts towards the west, with seep field emissions bypassing WCS. Given that an atmospheric plume survey can derive emissions in a couple of minutes, multiple seep areas should be repeat surveyed from morning through early evening to characterize the relationship between wind speed and emissions.

The study focus was marine seepage and neglected terrestrial seepage; however, the data analysis demonstrated that there are notable terrestrial emissions. Further WCS data analysis should assess emissions from Ellwood Field, with onshore surveys searching for seep emissions in neighborhoods by measuring seep gases in the near downwind of abandoned wells and faults, particularly those that intersect with the Ellwood Field.

Finally, the downcurrent dissolve<u>d</u> plume's fate is poorly understood as there is an absence of field data collected under typical afternoon winds. A combination of field data collected downwind of the downcurrent plume, ideally in conjunction with water sampling, and further modeling of WCS data can help constrain this emission source and better characterize the fate of COP seep field emissions.

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366 S8.2 Model improvements

367 The area plume inversion model was used to derive emissions on the field scale and sector scale. 368 Efforts to derive emissions at finer spatial scales were stymied by uncertainty in implementation of 369 wind veering. At the field scale, there was very low sensitivity of E to wind veering (Fig. S14); 370 however, simulations without wind veering shifted emissions off of important seeps like Trilogy Seep 371 to nearby seeps that are much smaller than Trilogy Seeps. Additionally, the model assigned emissions 372 to the field edges. Simulations suggested that wind veering that varies with wind direction is needed 373 - consistent with the field observations, i.e., wind veering for the southeast and southwest are different. This is not surprising given the different orientations of the coast on the east and west sides 374 375 of COP, as well as the topography between WCS and the coast - bluffs (~20 m) on the east side, and 376 their lack to the west (Sands Beach). Additionally, further offshore, wind veering likely becomes less 377 varying across the field and also shifts more towards prevailing. Further field observations are needed

to correctly characterize and incorporate wind veering to allow simulations to characterize changesto different areas of the field.

The model's enhancement of seepage at the seep field edges likely is from a combination of wind veering and outgassing emissions from downcurrent plumes. Currently, outgassing in the seep field area is assigned to the nearest seep area along the wind direction. This causes a problem for wind directions in the seep field with no seepage in that direction, including beyond the seep field edges.

Attempts to uniformly assign emissions with distance – i.e., no preferences – led to an unrealistic shift of emissions to the inshore seeps (based on a relative sonar return). A series of simulations where WCS was "shifted" north showed a linear dependency on emissions, and thus the model assigned emissions in any direction linearly with distance. One approach would be to prioritize major seepage over minor seepage in the other seep trend. Such an approach should follow a series of field emissions validation surveys.

390 There are a number of other model improvements that were not implemented due to 391 computational limits – current simulations take multiple days on a fast workstation (16-core 392 PowerMac, 96 GB RAM, 3.4 GHz). For example, rather than simulating the mean value of winds in 393 a direction, the probability distribution of u in each wind direction, e.g., Fig. 5A, could be used, 394 calculating essentially a weighted concentration for comparison with the observed concentration at 395 WCS. This would significantly increase computational load. An additional improvement would be to 396 include a boundary layer height that varied with wind speed and the diurnal cycle, and turbulence 397 parameterizations that are based on solar insolation. Values of boundary layer height and solar 398 insolation could be retrieved from NOAA weather models. It should be noted that the current model 399 complexity required several days to simulate at the highest spatial resolutions on a fast workstation. 400 Finally, higher angular resolution and sonar map resolution (doing one without the other causes non-401 physical striation) improves the ability to capture changes at different seep areas, but the increased 402 angular resolution introduces more gaps in the seep emissions source function. This could be 403 addressed by adding a "dispersed" source of sea air gas evasion that follows currents.

404

405 Supplemental References

Briggs, G. A.: Diffusion estimation for small emissions, NOAA, Air Resources Atmospheric
Turbulence and Diffusion Laboratory, Oak Ridge, Tennessee, 87-147, 1973.

- 408 Hanna, S. R., Briggs, G. A., and Hosker Jr., R. P.: Handbook on Atmospheric Diffusion, edited by:
- 409 Smith, J. S., Technical Information Center, U.S. Department of Energy, 110 pp., 1982.
- 410 Leifer, I., Kamerling, M., Luyendyk, B. P., and Wilson, D.: Geologic control of natural marine
- 411 hydrocarbon seep emissions, Coal Oil Point seep field, California, Geo-Marine Letters, 30, 331-338,
- 412 <u>https://doi.org/10.1007/s00367-010-0188-9</u>, 2010.
- 413 Leifer, I., Melton, C., Frash, J., Fischer, M. L., Cui, X., Murray, J. J., and Green, D. S.: Fusion of
- 414 mobile in situ and satellite remote sensing observations of chemical release emissions to improve
- disaster response, Frontiers in Science, 4, 1-14, <u>https://doi.org/10.3389/fenvs.2016.00059</u>, 2016.
- 416 Leifer, I., Chernykh, D., Shakhova, N., and Semiletov, I.: Sonar gas flux estimation by bubble
- 417 insonification: Application to methane bubble flux from seep areas in the outer Laptev Sea, The
- 418 Cryosphere, 11, 1333-1350, <u>https://doi.org/10.5194/tc-11-1333-2017</u>, 2017.
- 419