**Deciphering the variability in air-sea gas transfer due to sea state and wind history**

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**Supplement**

*Machine learning (ML) analysis of gas transfer observations*





Figure S1. Performance of the ML model for the training dataset (top) and testing dataset (bottom). The mean squared error (MSE in cm2 hr-2) and the coefficient of determination (R2) are labeled on top of the panels. Blue line represents 1:1, while the red line is linear regression. The Pearson correlation coefficients (r2) are 0.97 and 0.81 for the training and testing datasets, respectively.



Figure S2. Performance of the ML model for the additional validation dataset from Dong et al. 2024, which was not used to train the model. The ML model overpredicts relative to the observations by 14% (R2 = 0.66) but the Pearson correlations between observations and predicts remain high (r2 = 0.74).

To understand why the ML model can better predict *KCO2,660* than wind speed, we interrogate the factors contributing to the predictions from ML via the SHAP value. SHAP (SHapley Additive exPlanations) is a game theory approach to understand how the ML model makes predictions. This approach looks at all the input variables and evaluates which variables has the largest impact on the model output.



Figure S3 shows the input variables for the ML model along the Y-axis, with the variables ranked by how large their impact is on the model prediction. The X-axis shows the SHAP value, which is the scale of the output prediction from the model. Unsurprisingly we see that *U*10*n* is the most important variable for the ML model to make a prediction – the largest positive predictions of *KCO2,660* are for high *U*10*n* (in red), while the lowest predictions of *KCO2,660* are for low *U*10*n* (blue). Significant wave heights are also correlated with *KCO2,660* and show relatively large SHAP value, while inputs such as wave age and wave steepness have little impact on the predicted *KCO2,660*.

**ML model input parameters:**

*In situ*

U10n – In situ 10-m neutral windspeed

U10-1hr-lag – U10n from the previous hour

SST\_C – In situ bulk sea surface temperature

Latitude – Cos(latitude in degrees / 180 $π$)

*ECMWF (high resolution ECWAM) wave parameters (see Yang et al. 2022)*

Hs\_windsea – Significant wave height (windsea)

Hs\_swell – Significant wave height of (windsea)

Hs\_total – Significant wave height (total)

Wdir\_swell – Arctan of the difference between the wind direction and swell direction

MeanPeriod – Energy mean period of total waves

MeanPeriod\_swell – Energy mean period of swell

Total\_Wavelength – Wavelength from total wave period

Wave\_total\_steepness – Steepness of waves

Wave\_Cp – Phase speed of waves.

WaveAge – Wave age

WaveAge – Inverse wave age

*Calculations of wave parameters:*

Wavelength($λ$, in m) is computed from the energy mean period (Pm) of the total waves (in s) following the deep water dispersion relationship: $λ$ = *g* Pm 2 / (2 $π$), where *g* is gravity (m s-2). Total wave steepness is the ratio between the total significant wave height and $λ$. Wave age is computed as the ratio between the phase speed (*Cp*) of waves and *U*10*n*, where

*Cp* = (*g*/(2 $π$ / $λ$))0.5. Inverse wave age is simply 1/wave age.

*­Variability in significant wave heights (windsea, swell, total) as a function of wind history*



Figure S4. Bin-averages of significant wave heights over all datasets considered. Unsurprisingly, significant wave heights are on average greater during falling wind than during rising wind.

Table S1. Relative enhancement (%) in *KCO2,660* (all data, threshold of |0.5 m s-1 h-1|) and *Wf* (average of Knorr11 cruise and Callaghan et al. 2008) during conditions of falling winds vs. rising winds, in different wind speed bins.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | 7-9 m s-1 | 9-11 m s-1 | 11-13 m s-1 | 13 – 15 m s-1 | 15-17 m s-1 | 17-19 m s-1 | 19-21 m s-1 | 21-23 m s-1 |
| *KCO2,660* | 3 | 10 | 7 | 35 | 34 | 39 | 35 | 16 |
| *Wf* | 49 | 65 | 62 | 41 | 61 | 38 | 34 | - |

Table S2. Pearson correlation coefficients (r) between *KCO2,660*and wind history within different wind speed bins (number of hours italicized). Wind histories (d*U*10*n*/dt) is computed as the linear trend of the preceding 3 hours. Hs\_windsea, Hs\_swell, and Hs\_total indicate windsea, swell, and total contributions to significant wave height. Significant correlations (95% confidence) are bold faced. Correlations with wave heights are almost always positive and higher with increasing *U*10*n*. Correlations with wind history are generally negative.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | 2-3 m s-1 (*86*) | 3-4 m s-1 (*113*) | 4-5 m s-1 (*155*) | 5-6 m s-1 (*156*) | 6-7 m s-1 (*188*) | 7-8 m s-1 (*216*) | 8-9 m s-1 (*225*) | 9-10 m s-1 (*198*) |
| Hs\_windsea | 0.128 | 0.052 | 0.056 | 0.120 | **0.254** | **0.221** | -0.033 | 0.134 |
| Hs\_swell | **0.213** | **0.229** | 0.007 | 0.083 | 0.115 | **0.185** | **0.174** | **0.180** |
| Hs\_total | **0.217** | **0.223** | 0.024 | 0.090 | 0.133 | **0.217** | **0.154** | **0.201** |
| dU10n/dt | -0.211 | **-0.220** | **-0.173** | -0.091 | -0.128 | -0.030 | 0.011 | 0.113 |

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | 10-11 m s-1 (*211*) | 11-12 m s-1 (*166*) | 12-13 m s-1 (*123*) | 13-14 m s-1 (*104*) | 14-15 m s-1 (*83*) | 15-16 m s-1 (*58*) | 16-17 m s-1 (*42*) | 17-18 m s-1 (*27*) |
| Hs\_windsea | **0.144** | **0.166** | **0.338** | **0.269** | **0.371** | **0.318** | 0.258 | **0.463** |
| Hs\_swell | **0.298** | 0.103 | **0.453** | **0.604** | **0.457** | 0.247 | **0.619** | **0.458** |
| Hs\_total | **0.309** | 0.146 | **0.464** | **0.533** | **0.474** | **0.319** | **0.461** | **0.569** |
| dU10n/dt | -0.122 | -0.021 | -0.167 | -0.121 | **-0.365** | -0.078 | -0.090 | -0.355 |

Data from Dong et al. (2024a), filtered for air-sea CO2 fugacity difference of at least 20 µatm, supplemented here with wave data from ECMWF.

Dong, Y. et al. (2024a). Data from: Direct observational evidence of strong CO2 uptake in the Southern Ocean. Dryad. <https://doi.org/10.5061/dryad.b2rbnzspm>

Eddy covariance CO2 flux and transfer velocity data from other JCR ORCHESTRA cruises (TS19, TS20, JCR19001, JCR30001)

Missing fields noted by NaN

Data column descriptions:

YearMonthDay\_TimeUTC : timestamp for middle of hourly average

lat\_deg : decimal latitude

lon\_deg : decimal longitude

CO2flux\_mmol\_m2\_d : eddy covariance CO2 flux, mmol/m2/d

kCO2660\_cm\_hr : CO2 transfer velocity at Sc=660, cm/hr

kCO2\_cm\_hr : CO2 transfer velocity at ambient Sc

ustar\_insitu\_m\_s : friction velocity from in situ wind measurements, m/s

U10n\_insitu\_m\_s : 10-m neutral wind speed from in situ wind measurements, m/s

SST\_C : bulk underway water temperature, deg C

Tskin\_C : skin temperature (bulk temperature corrected by COARE3.5 model cool skin effect), deg C

sal : salinity (assumed to be 35 if missing)

dfCO2\_uatm : sea-air difference in fCO2 (=fCO2w - fCO2a), uatm

sol\_CO2\_bulk\_mole\_m3\_atm : CO2 solubility computed at bulk water temperature, moles/m3/atm

sol\_CO2\_skin\_mole\_m3\_atm : CO2 solubility computed at skin temperature, moles/m3/atm

DCCO2\_mmol\_m3 : sea-air difference in CO2 concentration (=sol\_CO2\_bulk\_mole\_m3\_atm\*fCO2w - sol\_CO2\_skin\_mole\_m3\_atm\*fCO2a), mmol/m3

Sc\_CO2\_bulk : Schmidt number of CO2 computed at bulk water temperature

U10n\_ECMWF\_m\_s : 10-m neutral wind speed from ECMWF model, m/s

ustar\_ECMWF\_m\_s : friction velocity from ECMWF model, m/s

Hs\_ECMWF\_tot\_m : significant wave height of total waves from ECMWF model, m

RHw0 : wave Reynolds number at 20 deg C computed from Hs\_ECMWF\_tot\_m and ustar\_insitu\_m\_s

MSS\_ECWAM : mean squared slope of waves from ECMWF model

Hs\_ECMWF\_windsea\_m : significant wave height of wind sea, m

Hs\_ECMWF\_swell\_m : significant wave height of swell, m

MeanPeriod\_ECMWF\_tot\_s : mean wave period of total waves, s

MeanPeriod\_ECMWF\_swell\_s : mean wave period of swell, s

MeanPeriod\_ECMWF\_windsea\_s : mean wave period of wind sea, s

Wdir\_ECMWF\_deg : wind direction from ECMWF model, deg

Swell\_dir\_ECMWF\_deg : swell direction from ECMWF model, deg

YearMonthDay\_TimeUTC, lat\_deg, lon\_deg, CO2flux\_mmol\_m2\_d, kCO2660\_cm\_hr, kCO2\_cm\_hr, ustar\_insitu\_m\_s, U10n\_insitu\_m\_s, SST\_C, Tskin\_C, sal, dfCO2\_uatm, sol\_CO2\_bulk\_mole\_m3\_atm, sol\_CO2\_skin\_mole\_m3\_atm, DCCO2\_mmol\_m3, Sc\_CO2\_bulk, U10n\_ECMWF\_m\_s, ustar\_ECMWF\_m\_s, Hs\_ECMWF\_tot\_m, RHw0, MSS\_ECWAM, Hs\_ECMWF\_windsea\_m, Hs\_ECMWF\_swell\_m, MeanPeriod\_ECMWF\_tot\_s, MeanPeriod\_ECMWF\_swell\_s, MeanPeriod\_ECMWF\_windsea\_s, Wdir\_ECMWF\_deg, Swell\_dir\_ECMWF\_deg

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2019-12-17 10:30:00, -54.3231, -46.972, -2.92223, 10.0867, 6.53416, 0.155125, 5.10882, 4.6904, 4.5611, 33.9848, -31.1822, 54.6212, 54.7034, -1.86942, 1581.36, 4.93456, 0.161758, 1.68898, 275443, 0.0189, 0.285, 1.66475, 7.56125, 7.717, 2.40405, 300.414, 300.553

2019-12-17 11:30:00, -54.2691, -47.2944, -4.6176, 13.0536, 8.434, 0.167275, 5.4214, 4.60597, 4.4863, 33.9572, -38.3416, 54.759, 54.8601, -2.28701, 1587.8, 4.90857, 0.163141, 1.72872, 281687, 0.018825, 0.279425, 1.70595, 7.6671, 7.8154, 2.31825, 294.928, 294.495

2019-12-17 12:30:00, -54.2159, -47.6191, -8.50215, 22.4519, 14.4887, 0.228725, 7.0254, 4.56682, 4.40085, 33.9466, -42.164, 54.9105, 55.0339, -2.45442, 1595.18, 5.58101, 0.176544, 1.79022, 333152, 0.019075, 0.357425, 1.75217, 7.7533, 7.97002, 2.72155, 262.924, 288.38

2019-12-17 13:30:00, -54.1659, -47.9479, -7.44262, 27.275, 17.5425, 0.3039, 8.71235, 4.45947, 4.32338, 33.9075, -32.3026, 55.0888, 55.2006, -1.77301, 1603.49, 7.23882, 0.238718, 1.90217, 478420, 0.020625, 0.613275, 1.7948, 7.68915, 8.16525, 3.618, 232.871, 281.428

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2019-12-19 08:30:00, -53.1437, -52.7661, -5.8676, 21.993, 14.8243, 0.425, 11.2008, 5.97927, 5.90483, 34.0041, -26.9365, 52.1195, 52.1202, -1.65355, 1459.26, 12.6093, 0.569938, 2.81305, 1.58918e+06, 0.04165, 2.23572, 1.70507, 6.57773, 7.9677, 5.81027, 292.165, 31.4694

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2019-12-19 13:30:00, -52.8244, -54.13, -12.1455, 32.58, 22.079, 0.418925, 11.0867, 6.15535, 6.04547, 33.9757, -41.9181, 51.8216, 51.8682, -2.29896, 1444.55, 10.6497, 0.435234, 2.8398, 1.23482e+06, 0.036175, 1.74527, 2.23717, 7.08948, 8.13645, 5.41482, 251.176, 342.86

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2019-12-19 17:30:00, -52.6469, -55.0283, -13.0617, 24.9953, 17.2735, 0.3965, 10.637, 6.79855, 6.64147, 33.915, -62.3591, 50.7065, 50.8046, -3.16205, 1390.54, 9.11932, 0.364366, 2.42148, 868373, 0.031875, 1.31393, 2.03255, 6.9768, 7.93022, 4.72133, 225.455, 337.509

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2019-12-19 19:30:00, -52.677, -55.1229, -13.7795, 25.695, 17.782, 0.294275, 8.53133, 6.84492, 6.67564, 33.9103, -63.3261, 50.6227, 50.7449, -3.24027, 1386.52, 8.02503, 0.308958, 2.316, 704735, 0.029525, 1.07742, 2.04883, 7.09057, 7.8285, 4.4387, 218.503, 337.856

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2019-12-20 02:30:00, -52.6557, -55.0594, -6.4667, 11.6402, 8.04577, 0.1867, 5.94942, 6.80492, 6.59911, 33.8964, -66.2946, 50.6951, 50.885, -3.36071, 1389.77, 5.78593, 0.204866, 2.3208, 473810, 0.026425, 0.492825, 2.26745, 7.50105, 7.714, 2.9874, 220.549, 330.268

2019-12-20 03:30:00, -52.6745, -55.1028, -7.27232, 12.9659, 8.96246, 0.2079, 6.49142, 6.80645, 6.61213, 33.88, -67.342, 50.7024, 50.866, -3.3932, 1389.92, 6.22488, 0.21363, 2.32457, 507882, 0.0265, 0.56085, 2.2536, 7.49455, 7.75385, 3.302, 228.746, 329.983

2019-12-20 04:30:00, -52.6906, -55.1443, -9.73522, 17.5634, 12.1434, 0.23265, 7.12643, 6.81452, 6.64307, 33.8837, -67.1839, 50.6835, 50.8098, -3.35223, 1389.07, 7.33959, 0.253426, 2.34035, 610135, 0.027075, 0.751125, 2.21177, 7.43843, 7.8429, 3.92248, 234.306, 332.623

2019-12-20 05:30:00, -52.7025, -55.1915, -14.5523, 26.2683, 18.1634, 0.29265, 8.46257, 6.81712, 6.65561, 33.892, -67.1211, 50.6774, 50.7853, -3.35017, 1388.89, 8.32781, 0.297721, 2.36655, 712889, 0.028125, 0.958075, 2.16065, 7.34015, 7.91113, 4.4312, 236.497, 334.315

2019-12-20 06:30:00, -52.7053, -55.22, -12.7804, 23.0021, 15.8963, 0.342225, 9.552, 6.79898, 6.65263, 33.891, -67.6369, 50.6989, 50.7909, -3.36112, 1389.89, 8.58402, 0.316507, 2.38978, 754183, 0.029125, 1.07308, 2.13443, 7.24812, 7.90808, 4.65027, 237.519, 327.438

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2019-12-20 09:30:00, -52.499, -55.8926, -10.0856, 14.861, 10.281, 0.1955, 6.1852, 6.834, 6.69844, 33.8969, -86.6917, 50.5921, 50.7078, -4.0976, 1384.9, 7.52348, 0.276414, 2.2503, 612085, 0.029, 0.8707, 2.07382, 7.0524, 7.58338, 4.06557, 252.91, 244.117

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2019-12-20 13:30:00, -52.1536, -57.059, 5.564, -10.3739, -7.32511, 0.170675, 5.55023, 7.51268, 7.27563, 33.8327, -62.9782, 49.6131, 49.7124, -3.18478, 1338.08, 5.54934, 0.197926, 1.73843, 335722, 0.0227, 0.424825, 1.68513, 7.00793, 7.2702, 2.8636, 304.534, 175.128

2019-12-20 14:30:00, -52.0815, -57.3507, -2.67105, 4.29107, 3.07116, 0.14165, 4.65022, 7.964, 7.74726, 33.7176, -77.5686, 48.8499, 48.9358, -3.64465, 1301.27, 5.22145, 0.176455, 1.59535, 281109, 0.020425, 0.342525, 1.55795, 7.07705, 7.29433, 2.58093, 314.351, 153.162

2019-12-20 15:30:00, -52.0142, -57.6354, -2.0587, 3.7082, 2.6664, 0.091325, 3.02645, 8.12053, 7.96577, 33.6583, -69.4194, 48.4687, 48.5837, -3.22658, 1283.03, 6.08101, 0.201913, 1.44, 301127, 0.018625, 0.371975, 1.3893, 7.03637, 7.36995, 2.5797, 315.893, 139.447

2019-12-20 16:30:00, -51.9746, -57.9078, -6.54117, 13.3662, 9.65072, 0.0725, 2.37498, 8.25908, 7.99593, 33.6193, -60.628, 48.3957, 48.5429, -2.84123, 1279.24, 7.36814, 0.274084, 1.2591, 353238, 0.017975, 0.465475, 1.16595, 6.69292, 7.38738, 2.64043, 319.735, 137.437

2019-12-20 17:30:00, -51.951, -58.1811, -5.19305, 5.22029, 3.85306, 0.153825, 4.50985, 9.00387, 8.68845, 33.5928, -111.873, 47.2808, 47.4148, -5.6602, 1228.17, 8.19898, 0.327834, 0.994775, 333012, 0.0184, 0.529925, 0.8301, 5.8795, 7.33632, 2.68995, 323.258, 136.065

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2020-01-13 11:30:00, -67.567, -68.2256, -0.872375, 0.854754, 0.495899, 0.05075, 1.52842, 1.19305, 1.05326, 32.4277, -122.887, 62.7838, 62.8719, -7.35912, 1973.83, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN

2020-01-13 12:30:00, -67.567, -68.2258, -0.1004, 0.098714, 0.0572895, 0.0683, 2.15862, 1.20348, 1.06453, 32.4464, -122.174, 62.7352, 62.8386, -7.32916, 1971.61, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN, NaN

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2020-11-27 17:30:00, -37.5062, -31.7528, -0.211775, 0.571284, 0.523232, 0.251775, 7.58812, 16.8, 16.7222, 35.5648, -39.6542, 36.2653, 36.3909, -1.68444, 785.182, 8.98192, 0.361694, 3.17362, 1.10735e+06, 0.033675, 1.37902, 2.82673, 7.77003, 8.42865, 4.94505, 261.474, 298.041

2020-11-27 18:30:00, -37.6709, -31.7908, -2.83665, 6.4517, 5.88571, 0.29755, 8.61397, 16.6482, 16.5426, 35.6027, -45.2248, 36.467, 36.5784, -2.00831, 793.15, 8.03509, 0.306257, 3.14893, 952718, 0.0312, 1.08432, 2.95503, 8.03843, 8.5558, 4.41277, 258.729, 292.938

2020-11-27 19:30:00, -37.836, -31.8322, -3.43785, 7.96027, 7.27808, 0.32895, 9.28745, 16.7334, 16.6174, 35.5995, -45.0122, 36.3884, 36.4979, -1.96901, 790.12, 7.95542, 0.294107, 3.12328, 914870, 0.0298, 1.03392, 2.946, 8.18885, 8.68923, 4.3398, 265.469, 289.703

2020-11-27 20:30:00, -37.9996, -31.8736, -6.1638, 14.7843, 13.5043, 0.33645, 9.44475, 16.6964, 16.6081, 35.5935, -44.4714, 36.4001, 36.509, -1.90109, 790.529, 8.02729, 0.294564, 3.10907, 916436, 0.029325, 1.02175, 2.9352, 8.23682, 8.73745, 4.31465, 267.545, 287.369

2020-11-27 21:30:00, -38.1626, -31.9145, -7.70975, 19.7823, 18.0576, 0.376575, 10.2568, 16.671, 16.5727, 35.5626, -39.3872, 36.4451, 36.5528, -1.77891, 792.046, 8.42259, 0.306225, 3.10993, 960608, 0.0295, 1.09608, 2.90815, 8.21482, 8.76925, 4.48635, 264.793, 286.049

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2020-11-27 23:30:00, -38.4919, -31.9904, -9.27305, 24.3444, 22.1984, 0.3556, 9.82467, 16.6303, 16.5502, 35.5543, -37.243, 36.4312, 36.5786, -1.73761, 791.456, 8.85382, 0.330462, 3.1405, 1.02907e+06, 0.030525, 1.3238, 2.84617, 8.1214, 8.82807, 4.9802, 253.739, 284.563

2020-11-28 00:30:00, -38.6533, -32.0314, -12.3105, 28.4722, 25.756, 0.413625, 10.9691, 16.3255, 16.2147, 35.57, -42.7422, 36.7958, 36.9428, -1.99014, 805.572, 8.66125, 0.320623, 3.16502, 1.00861e+06, 0.030625, 1.3155, 2.87723, 8.13748, 8.8205, 5.00432, 251.013, 281.869

2020-11-28 01:30:00, -38.805, -32.0677, -11.0266, 23.6167, 21.3208, 0.424225, 11.1735, 16.2488, 16.0828, 35.6094, -49.3908, 36.931, 37.0816, -2.15684, 811.058, 8.77802, 0.319584, 3.24433, 1.04018e+06, 0.03105, 1.41035, 2.91797, 8.1801, 8.88978, 5.23793, 244.887, 279.216

2020-11-28 02:30:00, -38.9468, -32.1019, -13.58, 29.6698, 26.7528, 0.43025, 11.2834, 16.2022, 16.0987, 35.6321, -48.0214, 36.8978, 37.0599, -2.11226, 809.933, 9.57353, 0.345893, 3.41307, 1.20834e+06, 0.032325, 1.77252, 2.90548, 8.22222, 9.0746, 5.95158, 237.333, 278.814

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2020-11-28 09:30:00, -39.8776, -32.3424, -2.63303, 5.76391, 4.9991, 0.237, 7.20825, 14.7391, 14.7308, 35.093, -39.3058, 38.5073, 38.7267, -2.18243, 869.074, 7.0036, 0.256392, 3.92115, 982368, 0.02755, 0.907125, 3.80997, 9.5474, 9.88377, 4.15415, 234.135, 253.801

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2020-12-13 19:30:00, -54.4912, -54.3819, -1.25022, 3.88249, 2.63006, 0.08425, 2.77995, 6.1423, 6.07924, 34.0302, -28.3497, 51.574, 51.7918, -1.97667, 1433.34, 4.60069, 0.163936, 2.88845, 456062, 0.023625, 0.3322, 2.86765, 8.84828, 8.95478, 2.37378, 304.058, 250.263

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2020-12-13 22:30:00, -54.8998, -53.911, -2.60255, 5.5909, 3.48308, 0.175, 5.66818, 3.43972, 3.37796, 33.7865, -47.452, 57.0834, 57.1874, -3.11478, 1701.86, 3.96791, 0.1235, 2.66393, 337888, 0.020475, 0.18705, 2.6568, 9.12343, 9.172, 1.98505, 8.14777, 255.784

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