

SUPPORTING INFORMATION

Global concentrations of gaseous elemental mercury and reactive gaseous mercury in the marine boundary layer

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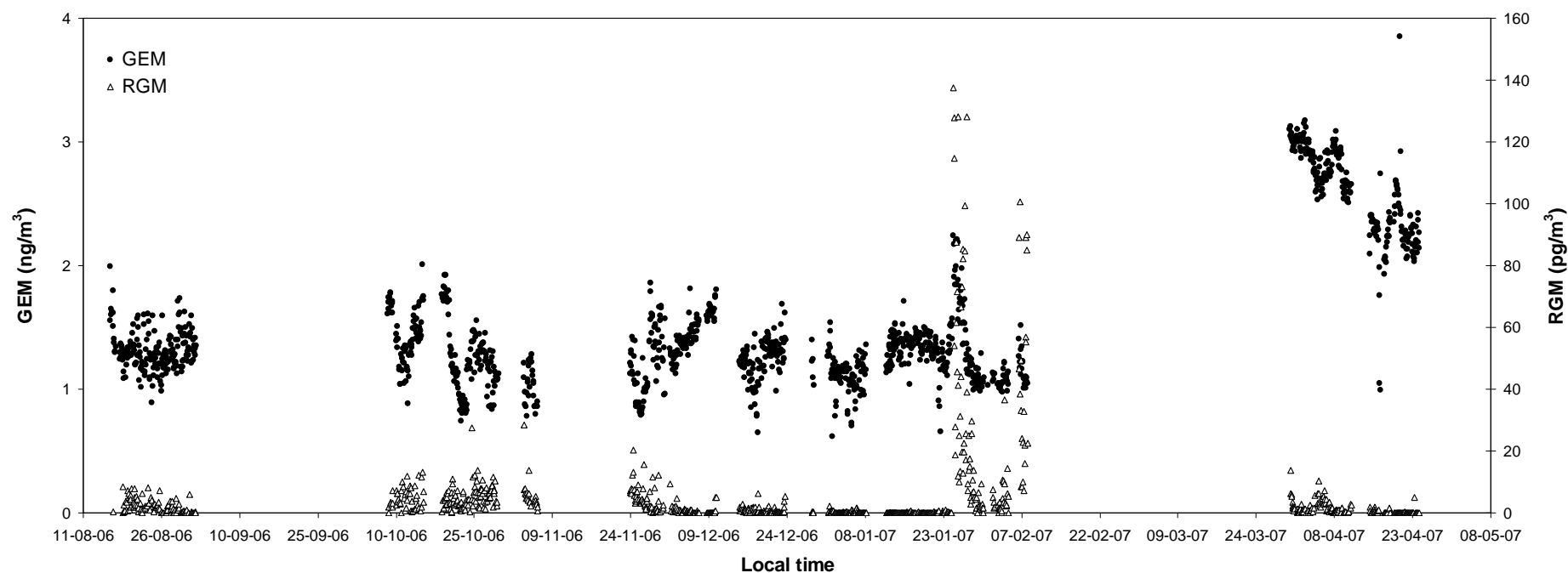
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Figure S1. 2 hour average GEM and RGM concentrations during the cruise

Two hour mean values for GEM and RGM during the cruise. Only measurements in the MBL are presented, while measurements from harbor areas are excluded and will be presented in another paper that is currently in review.



Text T1. Placement of instruments

All measurements were carried out from a 10' shipping container located on the port side of the ship next to the chimney and behind the bridge. The inlet was 8 meters above sea-level. The place was chosen to minimize the effect of ship emissions and Sea Spray.



Picture showing the location of the inlet for the Tekran (red circle) used and showing the inside of the thermostated container where the instruments were located.

Text T2. Measurement of RGM

RGM is a method defined parameter and thus the method has to be considered as a reference method, where the reproducibility of the method gives the best estimate of the uncertainty. In an inter-calibration exercise carried out at Svalbard in 2003 (1) the uncertainty of RGM measurements was found to be very large; agreement was at best to within an order of magnitude. Participating laboratories used a Tekran 2537A equipped with a Tekran 1130/5 sampling unit.

We carried out parallel measurement of RGM at Barrow, Alaska, where one group used a Tekran 2537A equipped with a Tekran 1130/5 sampling unit and the other only a Tekran 2537A equipped with 1130 (unpublished results). Because of the use of different sampling cycles of different duration, a direct comparison of measurements was not possible but 3 day averages showed very good agreement 1.7 % (one std. dev.). In Skov et al. (2) we compared a Tekran 2537A equipped with the Tekran 1130 automated system with manual denuders (3). Manual and automated methods were found to agree to within 25% (95% confidence interval). However we do not have sufficient data from the literature or from our own experiments to calculate the

reproducibility for RGM measurements on the Galathea 3 cruise. Moreover RGM is probably not made of the same gaseous Hg^{II} species but may differ in time and space. Therefore we added 10% to the uncertainty to make a conservative estimate. The expanded reproducibility of RGM is thus 28 %, using a factor of 2 to get a 95% confidence interval.

Text T3. Measurements of NOx and soot

NOx ($\text{NO} + \text{NO}_2$) was measured with an API Teledyne NOx analyzer Model 200 A. To ensure the quality of the NOx data zero-air was added frequently ($n=124$) and samples ($n = 10$) from a pressurized flask with a known, certified NO concentration were added every four weeks. NOx (NO and NO_2) is used as a tracer for contamination from the ship's exhaust. The uncertainty of the method is 4% for NO and 10 % for NO_2 (based on uncertainty budgets and using an expanding factor 2 following the guidelines in CEN EN 14211) for values above 1 ppbv. The detection limits are 200 and 300 pptv for NO and NO_2 respectively for the experimental setup applied. This relative high detection limit for the method is caused mainly by the quality of zero air and not by the capability of the instrument.

Soot data were only used for the (SA) leg during the passage along the Africa coast. Soot for this part of the cruise was made available by Matthias Ketzel (NERI, Aarhus University). The measurements were performed using a **P**article **S**oot **A**bsorption **P**hotometer (PSAP). A description of the instrument can be found in Krecl et al. (4). We used a median of 2 hour mean soot data ($n=13$) to be consistent with other data in the paper. Quality control checks were made on the 5-minute-interval measurements. The ship was moving against the wind for all the measurements along the African coast and we did not see indications of contamination from the ship.

Table Ta1. Availability of supplementary measurements of NO_x, soot and meteorological data

Leg	Ab ^a	Dates	Lat ^b	Origin ^c	NO Mean (ppb)	NO Median (ppb)	NO ₂ Mean (ppb)	NO ₂ Median (ppb)	Soot ^e Mean (m ⁻¹)	RH Mean (%) (no. obs) ^d	Radaition Mean midday (11-13, local time) (W/m ²) (no. obs) ^d
North Atlantic	NA	16 th Aug – 1 st Sep	58 : 67	Ocean						92.5±6.1 (185)	
Atlantic O.	AT	15 th Apr – 24 th Apr	43 : 59	Ocean	13.9±34.8	0.2	1.8±3.6	0.2		90.4±5.6 (113)	339±160 (9)
Sargasso Sea	SS	30 th Mar – 11 th Apr	23 : 45	Mixed	11.8±24.8	3.0	4.2±3.7	3.5		62.6±7.1 (142)	813±150 (12)
South Africa	SA	8 th Oct – 21 st Oct	-39 : -3	Mixed					1.5*10 ⁻⁶	86.3±6.7 (120)	498±198 (10)
Indian Ocean	IO	22 nd Oct – 29 th Oct	-39 : -33	Ocean						80.8±12.3 (91)	617±217 (8)
West Australia	WA	3 rd Nov – 6 th Nov	-22 : -17	Ocean						72.1±6.2 (32)	907 (2)
East Australia	EA	23 rd Nov – 15 th Dec	-44 : -26	Mixed	32.7±76.3	1.8	2.1±8.5	0.0		71.6±10.2 (186)	782±205 (15)
Coral Sea	CS	16 th Dec – 3 rd Jan	-27 : -7	Mixed	77.1±131.4	10.1	5.5±8.2	1.1		76.1±5.8 (129)	603±287 (11)
New Zealand	NZ	3 rd Jan – 14 th Jan	-56 : -26	Mixed	18.3±49.9	1.1	0.7±4.4	0.0		83.0±13.0 (109)	493±258 (9)
Antarctic O.	AO	14 th Jan – 24 th Jan	-65 : -55	Ocean	27.4±76.5	0.6	0.64±4.1	0.0		93.1±3.7 (106)	235±133 (9)
Antarctic Coast	AC	25 th Jan – 28 th Jan	-65 : -63	Ocean	7.0±15.9	1.5	0±2.0	0.0		92.5±4.0 (49)	415±176 (4)
Coast of Chile	CC	31 st Jan – 8 th Feb	-58 : -33	Mixed	90.2±157.2	15.13	3.15±10.2	0.1		63.8±11.2 (37)	638±407 (4)

a) Abbreviations used in the text.

b) The latitude that the given leg includes.

c) Ocean = no influence from terrestrial sources, Mixed = influence from terrestrial sources during the entire or smaller but significant portions of the leg.

d) The two hour means are used to indicate the number of observations of each species or parameters.

e) Soot measurements were performed by an independent group and we therefore only have limited access to results.

Table Ta2. Previous measurements in the MBL that overlap spatially with measurements during the cruise presented in the paper

Place	Date	TGM ng/m ³	Reference
Southern Hemisphere			
South Atlantic Ocean	Oct 77	1.19±0.25	Slemr et al. 1996
South Atlantic Ocean	Nov-Dec 78	1.35±0.21	Slemr et al. 1996
South Atlantic Ocean	Jan-Feb 79	1.26±0.22	Slemr et al. 1996
South Atlantic Ocean	Oct-Nov 80	1.45±0.16	Slemr et al. 1996
South Atlantic Ocean	Oct-Nov 90	1.50±0.30	Slemr et al. 1996
South Atlantic Ocean	Oct-Nov 94	1.18±0.17	Slemr et al. 1996
South Atlantic Ocean	Dec 99-Jan 00	1.27±0.09	Temme et al. 2003b
South Atlantic Ocean	Feb-Mar 00	1.00±0.12	Temme et al. 2003b
South Atlantic Ocean	Feb 01	1.07±0.10	Temme et al. 2003b
Antarctic – South America	Dec 01 – Feb 02	1.1±0.2	Temme et al 2003a
Indian Ocean	Nov 07	1.2 (1.05-1.51)	Witt et al. 2010
Northern Hemisphere			
Eastern Atlantic Ocean	Oct 77	1.76±0.36	Slemr et al. 1996
Eastern Atlantic Ocean	Nov-Dec 78	1.85±0.31	Slemr et al. 1996
Eastern Atlantic Ocean	Jan-Feb 79	2.17±0.38	Slemr et al. 1996
Eastern Atlantic Ocean	Oct-Nov 80	2.09±0.35	Slemr et al. 1996
Eastern Atlantic Ocean	Oct-Nov 90	2.25±0.41	Slemr et al. 1996
North Atlantic Ocean	Aug 93	2.1±0.8	Mason et al. 1998
Atlantic Ocean	Oct-Nov 94	1.79±0.4	Temme et al. 2003b
Eastern Atlantic Ocean	Oct-Nov 96	2.12±1.0	Temme et al. 2003b
Western Atlantic Ocean	Sep 99	2.00±0.4	Mason et al. 2001
Eastern Atlantic Ocean	Dec 99-Jan 00	2.02±0.3	Temme et al. 2003b
Western Atlantic Ocean	Aug-Sep 03	1.63±0.08	Laurier and Mason 2007
North Atlantic Ocean	July 05	~1.7 ^A	Sommar et al. 2009

A. Arctic data not including

B. Slemr et al. 1996 (5), Mason et al. 1998 (6), Mason et al. 2001 (7), Temme et al. 2003a (8), Temme et al. 2003b (9), Laurier and Mason 2007 (10), Sommar et al. 2009 (11), Witt et al. 2010 (12)

Text T4. Seasonal cycle in the Atlantic Ocean

The seasonal cycle in the Northern Hemisphere Atlantic Ocean is found by converting the available cruise data from the Northern Hemisphere Atlantic Ocean into monthly mean values.

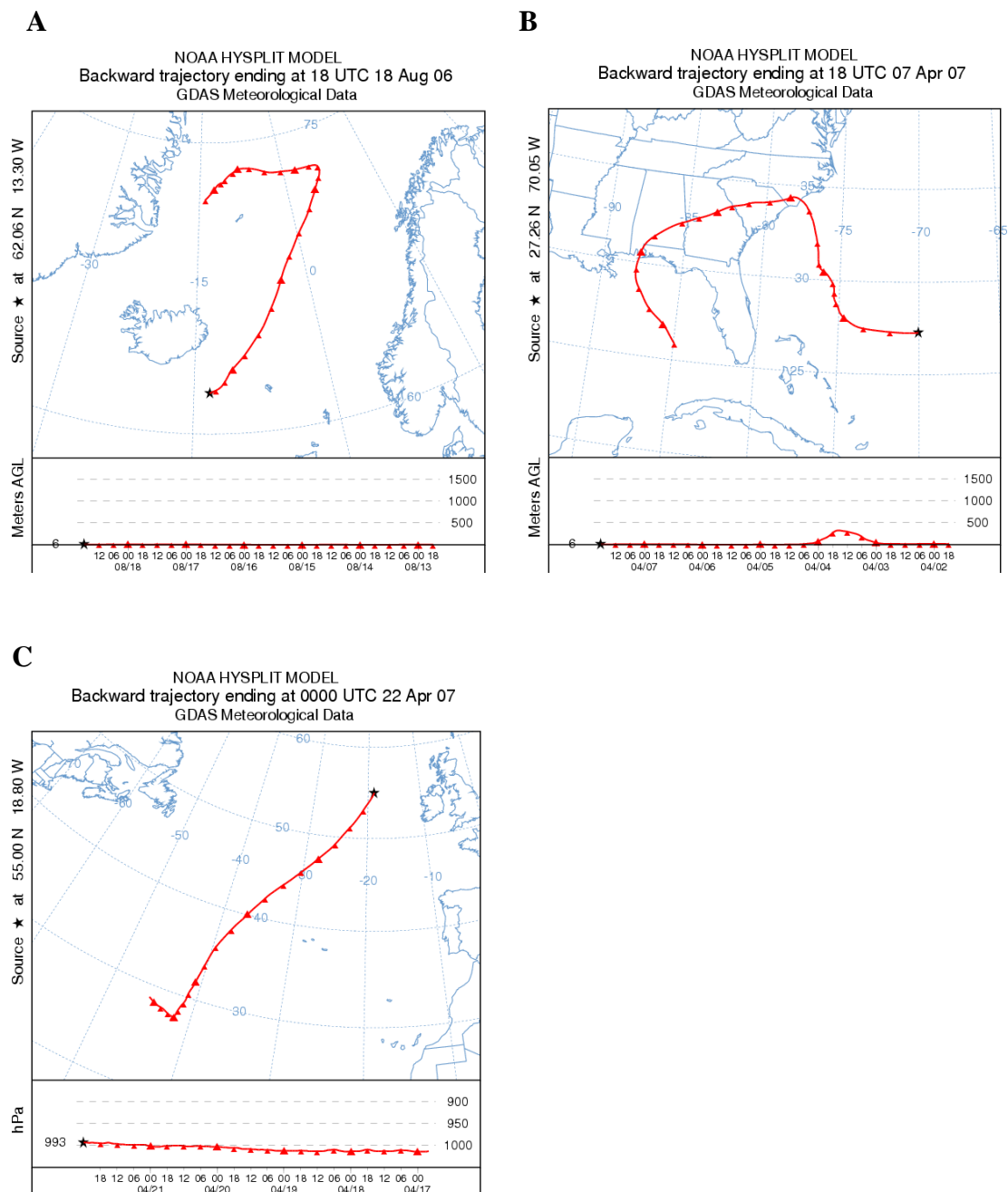
Mean values and standard deviations have been taken from earlier publications (6,7,9-11,13,14). When cruises had a duration of more than one month the same mean value was used to represent all months of the cruise except in the cases where information was sufficient to calculate means for the different months. This approach causes all cruises to have the same weight in the calculations despite their differences in duration and spatial coverage. This is a rough approximation but necessary at this point due to the sparse observations.

To create a smoothed line for the seasonal cycle the mean of the previous and following month were used to make a central moving average. No observations are available in May in the Atlantic Ocean and a value is estimated from the mean concentration in April and June.

Figure S2. Back trajectories in the Atlantic

The origin of a given air mass arriving to *Galathea 3* was determined throughout the campaign using back trajectory calculations. Below are shown three examples for the Atlantic Ocean; A) Norway – Greenland in August 2006, B) US Virgin Islands – Boston in March/April 2007, C) Boston – Copenhagen in April 2007.

HYSPLIT back trajectories were used from <http://ready.arl.noaa.gov/HYSPLIT.php> (15). The heights of the trajectories were 6 meters above sea level, the duration 144 hours.



HYSPLIT back trajectories were used from <http://ready.arl.noaa.gov/HYSPLIT.php> (15).



Figure S4. South Atlantic wind directions

While back trajectories for Africa are less clear, the wind direction measured at the ship, together with soot data, satellite CO data and fire counts, gave a good indication of potential mercury sources on the west coast of Africa. We think that this discrepancy between back trajectories and other data might be due to a strong sea-land breeze close to the African coast that is not caught by the HYSPLIT model.

The figure shows wind direction and GEM concentrations encountered along the coast of Africa. Case A: biomass burning, Case B: free MBL air. The red area was influenced by biomass burning (16) (see Figure S6) and the yellow area was influenced by the plume from biomass burning identified by an elevated CO column concentration on October 12th, 2006 (17) (see Figure S5).

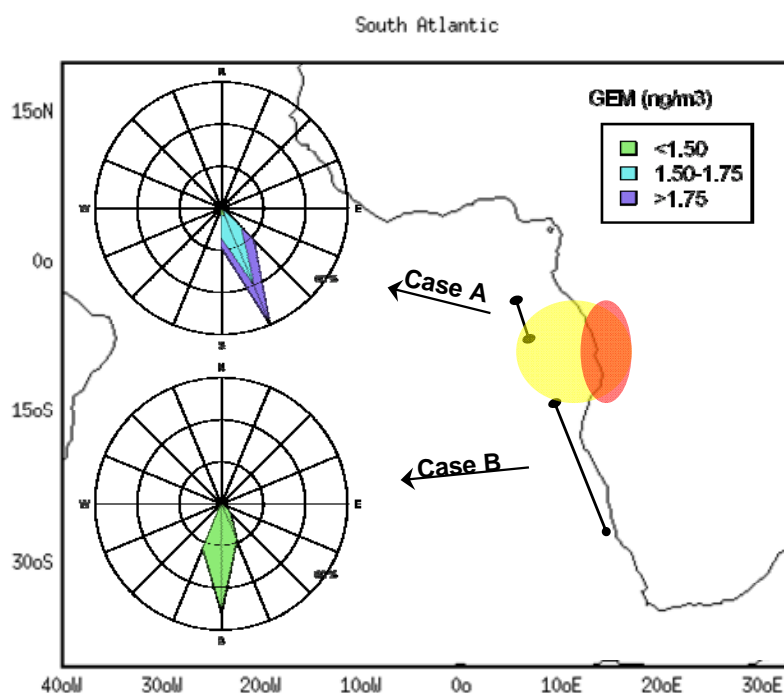


Figure S5. CO MOPIT data

Mean atmospheric CO columns (October 12th, 2006). MOPITT (V3) satellite observations obtained by the MOPIT Satellite (17).

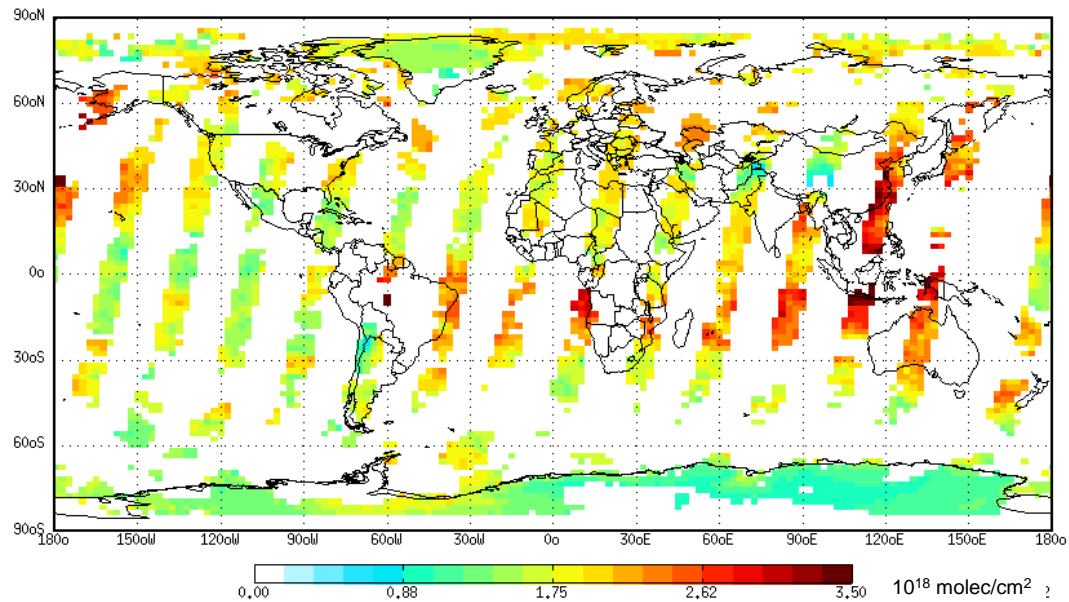


Figure S6. Fire counts in Africa during October 5th to 12th (2006).

Screen print from <http://firefly.geog.umd.edu/firemap/> (web fire mapper, accessed November 2009) (16).

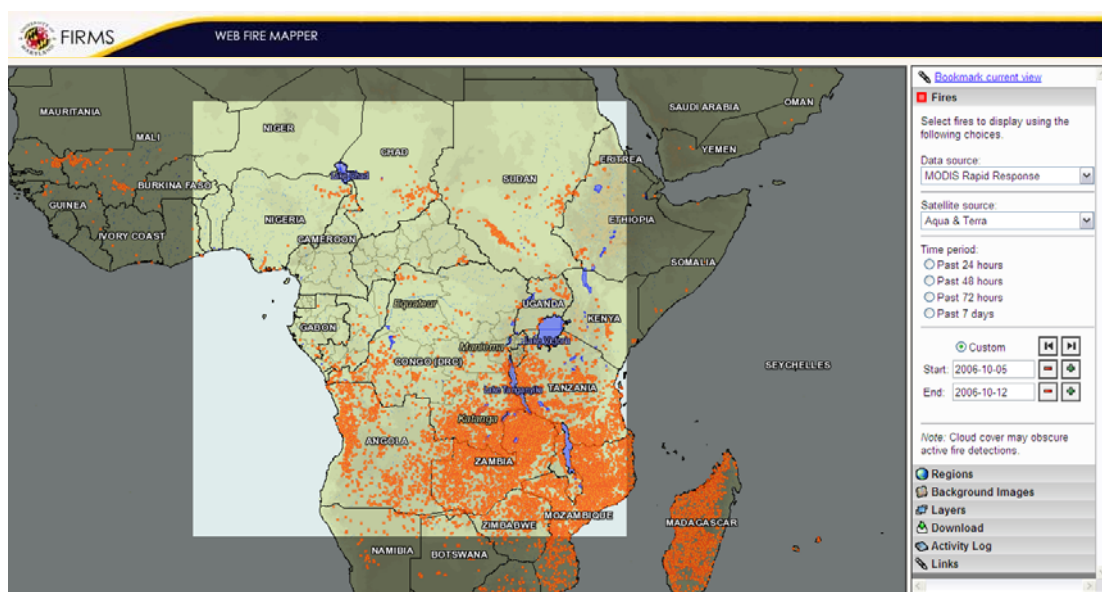


Figure S7. Back trajectories in the Antarctic

Back trajectories are from <http://ready.arl.noaa.gov/HYSPLIT.php> (15).

The heights of the trajectories are 6 meters above sea level, the duration is 96 hours.

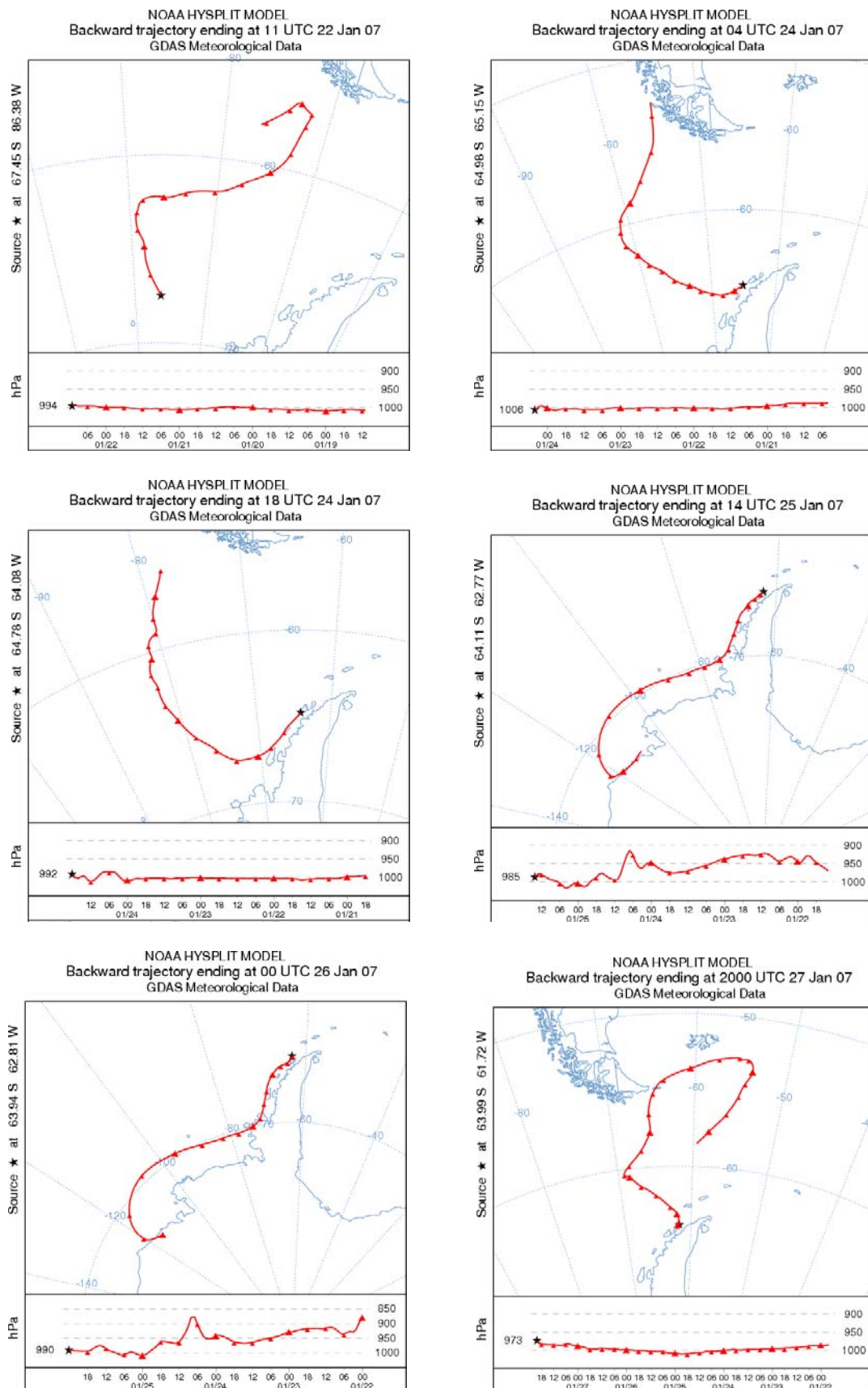
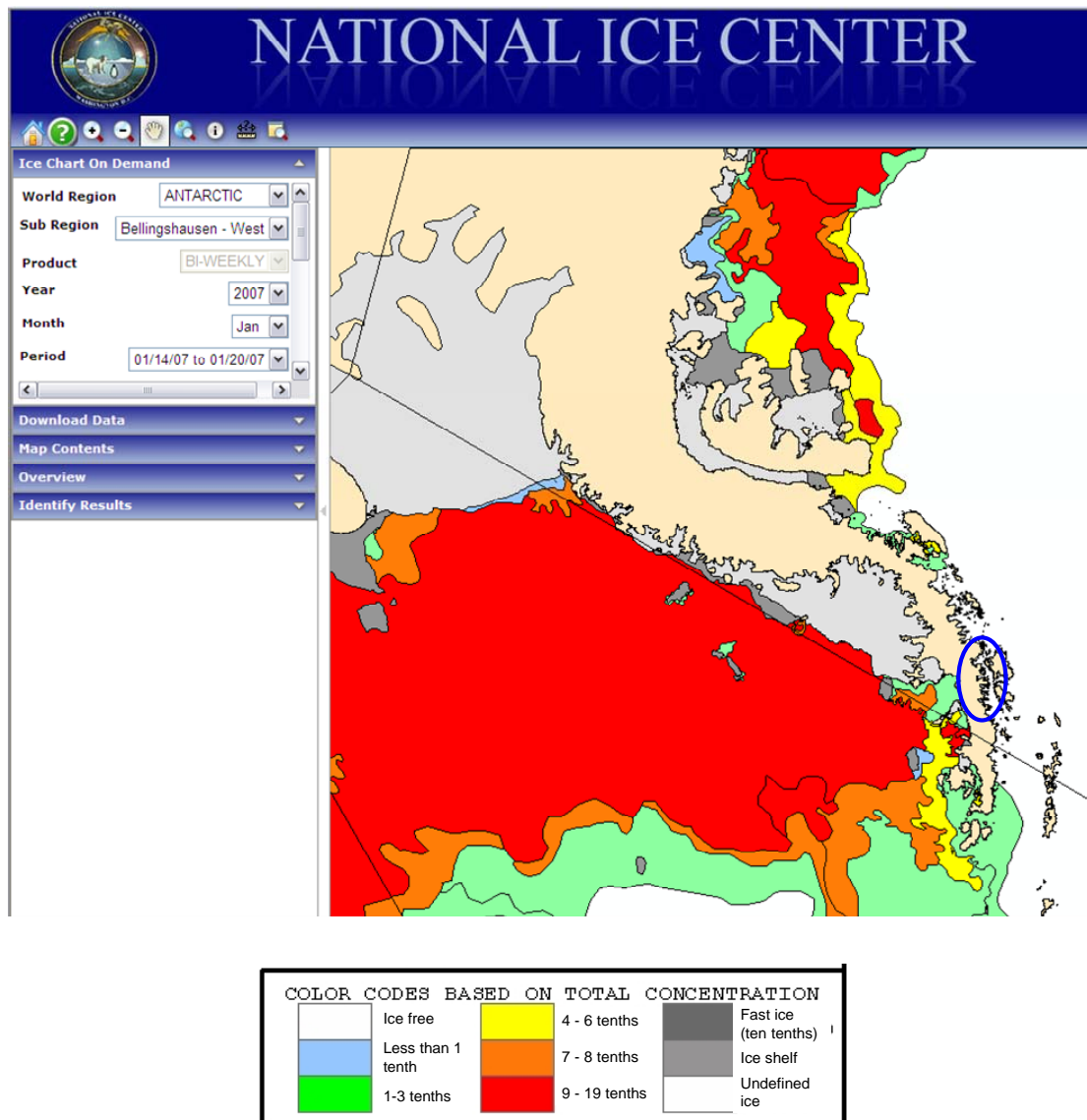


Figure S8. Ice extend (source: www.natice.noaa.gov, accessed December 2009)

Sea ice extend determined for the 14-20 January 2007.

Screen print of the ice extent at Bellingshausen West (included is a color scale that indicates the fraction of sea covered with ice). The blue circle indicates the location of the ship during the observations with high GEM and RGM concentrations.



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