

## Supplementary material

### Methods

#### *Dissolved trace metals*

Water column samples were collected using externally-closing, Teflon-lined Niskin-1010X bottles on an autonomous 1018 intelligent rosette system, specially adapted for trace metal work, and suspended on Kevlar rope (General Oceanics Inc., USA) [Ellwood *et al.*, 2008]. Dissolved iron (dFe) was determined at sea on acidified, 0.2  $\mu\text{m}$  filtered samples using a modification of a flow injection – chemiluminescence method (8  $\mu\text{M}$  final sulfite concentration was used for Fe(III) reduction over 1 h) [Bowie *et al.*, 1998, 2004; Hopkinson and Barbeau, 2007], which had been quality controlled against iron reference standards collected during the SAFe (“Sampling and Analysis of Fe”) project [Johnson *et al.*, 2007; data reported in Lannuzel *et al.*, submitted]. We also carried out an intercalibration for dFe on replicate samples collected during the SAZ-Sense voyage with a competitive ligand equilibration - adsorptive cathodic stripping voltammetry (CLE-ACSV) method following ultra-violet digestion [Croot and Johansson, 2000; Ibisanni *et al.*, submitted]. Consistency was excellent on 0-1000 m profiles at all three process stations, with differences insignificant at the 98% confidence interval (n=35) (data reported in full in Lannuzel *et al.* [submitted]).

#### *Particulate trace metals*

Suspended particulate trace metal samples were collected in situ using a 142 mm diameter stack consisting of a 54  $\mu\text{m}$  polyester pre-filter screen (Sefar Filtration, Inc.) followed by a Whatman QMA filter (nominal pore size 1  $\mu\text{m}$ ) supported by a 70  $\mu\text{m}$  Teflon PFA grid. Filter stacks were housed in large volume McLane pumps (McLane Research Laboratories Inc., USA) suspended on plastic-sheathed hydrowire. Filters and supports were prepared and pre-cleaned as described in Bishop *et al.* [1985]. Upon recovery of the pumps, filters were dried under a laminar flow bench

and sub-samples taken using a circular plastic punch (19 mm diameter). Filters were stored at ambient temperature in acid-washed petri dishes until further analysis in the home laboratory. Sinking particulate samples were collected in four separate cups, two each opened for 12 h on days 1 and 4 over the ~1 week long process stations, using a PPS3/3 free-floating sediment trap (Technicap, France) specially adapted for trace metals which was deployed at 150 m depth (full details of the trap deployment are given in *Ebersbach et al.* [submitted]). Upon recovery, particles collected in the cups were filtered off-line onto a 47 mm diameter, 2 µm porosity polycarbonate filter under gentle vacuum using a Teflon PFA unit (Savillex Corp., USA), equipped with a 1 mm porosity Teflon PFA pre-screening grid to exclude zooplankton ‘swimmers’. Blanks for both suspended and sinking particles were true ‘field’ blanks that were deployed in situ (i.e., filter stacks that no seawater had passed across on the McLane pumps, and cups that remained closed on the PPS3/3 sediment trap).

Sampled particles were acid extracted (1 ml concentrated HNO<sub>3</sub> for 4 h at 120 °C) in Teflon PFA vials and analyzed by magnetic sector inductively coupled plasma mass spectrometry (Finnigan ELEMENT 1) offering high spectral resolution capability, following *Cullen and Sherrell* [1999] and *Townsend* [2000]. Recoveries from the analysis of certified reference materials for marine sediments (National Research Council of Canada MESS-3 and PACS-2) and plankton (Community Bureau of Reference BCR-414) were approximately 70% for particulate iron (pFe) which mainly consisted of material of biogenic origin and some labile lithogenic fractions (probably from marine sediments) [*Lannuzel et al.*, submitted]. The low recovery of titanium (<10 %) in our certified reference materials suggest that the majority of lithogenic material was not easily digested. The missing fraction was likely a mixture of refractory lithogenic particles and biogenic (opal) material. Suspended biogenic opal represented a significant part of the total biogenic pool in a deep silica maximum and in the sinking particles collected with the PPS3/3 trap at station P2 (polar waters), but was a relatively minor fraction of both suspended and sinking particles at stations P1 and P3 (northern SAZ) [*Ebersbach et al.*, submitted; *K. LeBlanc*, personal

communication]. Thus our procedures probably measured "easily leachable" particulate biogenic iron (with a smaller lithogenic component). It is important to note that a variety of different sampling, filtration and analytical methods are currently used for particulate trace metals, which makes comparisons between datasets difficult, and there is currently no international standard for trace metals in suspended marine particles.

### *Iron uptake and remineralization*

Column integrated iron uptake rates (24 h deckboard incubations) and Fe/C cellular uptake ratios (based on  $^{55}\text{Fe}$  and  $^{14}\text{C}$  incorporation) were quantified for the resident phytoplankton and bacterial communities (i.e.,  $>0.2\ \mu\text{m}$ ) using radiotracer experiments following methods reported in *McKay et al.* [2005]. Samples were obtained from 6 depths (based on a range of water column light levels (50 to 1 % incident irradiance) collected using the trace metal rosette, and incubated at these light levels. Column integrated iron remineralization rates (based on 72 h in situ incubations in darkness) were estimated for the heterotrophic bacterial mobilization of algal iron from the  $>5\ \mu\text{m}$  size class only, following  $^{55}\text{Fe}$  radioisotope methods reported in *Boyd et al.* [submitted], and extrapolated to cells  $>0.2\ \mu\text{m}$  in size using the ratio of iron uptake-to-remineralization. This approach will underestimate iron remineralization by 3-4 fold as it does not include remineralization by bacterivory or herbivory [*Boyd et al.*, submitted] (see caption to Figure 4).

### *Aerosols*

Atmospheric sampling was conducted with an aerosol collector located on the deck above the bridge and forward of the exhaust stack. The aerosol collector was designed to cumulatively collect aerosols during several segments of cruise track by protecting the samples during non-sampling periods, thus allowing collection of small volume samples with concentrations above detection limits in remote ocean regions. This type of aerosol sampler has been deployed successfully on previous oceanographic cruises (see *Wagener et al.* [2008] for full details of cleaning and sampling

protocols). Eight samples (labeled AeroSAZ-1 to AeroSAZ-8) were collected along the entire cruise track over 35 days (Figure S1), with all samples corresponding to approximately 20 m<sup>3</sup> of filtered air (2-3 days of integrated sampling time). The aerosol total composition of iron and aluminium was determined by X-ray fluorescence following *Wagener et al.* [2008]. The aerosol iron dissolution in seawater was determined on separate filters following a “leaching” protocol similar to that described by *Buck et al.* [2006] and *Sedwick et al.* [2005]. It corresponds to an “instantaneous” solubility of iron that is obtained by quickly ( $\sim 2 \text{ ml s}^{-1}$ ) passing 60 ml of 0.2  $\mu\text{m}$  filtered surface seawater (collected from 15 m depth at P2,  $d\text{Fe} = 0.2 \text{ nmol l}^{-1}$ ) through the polycarbonate membrane used to collect the aerosols. The filtrate was directly collected in 60 ml vials and immediately acidified to pH 2 with HCl (Merck Ultrapur). This protocol was made successively twice on each aerosol filter. Dissolved iron in the “filtrate” was determined by flow injection – chemiluminescence following protocols and instrumentation described in *Blain et al.* [2008]. The amount of iron that dissolves is expressed as a percentage of total iron measured on the aerosol samples.

In order to provide a qualitative indication of preferential dust sources to the sampled areas, back trajectories of hypothetical air masses were calculated using the HYSPLIT (HYbrid Single Particle Lagrangian Integrated Trajectory) model developed at the NOAA Air Resource Laboratory [*Draxler and Rolph, 2003*], with re-analyzed archived meteorological data from the Global Data Assimilation System. Trajectories were hindcast at four different heights within the atmospheric boundary layer (10, 100, 500 and 1000 m) up to 120 h back in time. For each sampling day, a trajectory was calculated with a finishing point at the position of the ship at 12:00 (UTC) (Figure S1). Moreover, from September 2004 to February 2007, trajectories were calculated at 100 m up to 240 h back in time for every 24 h for the three finishing points at P1, P2 and P3. Even though errors on 10 day back trajectories may be significant [*Draxler and Rolph, 2003*], they provide a reasonable qualitative indication of preferential dust sources to the SAZ-Sense region.

### *Particulate organic carbon*

For POC analyses, the McLane large-volume pump QMA quartz filters (134 mm diameter, 1  $\mu\text{m}$  nominal pore size) were sub-sampled in a flow-bench with a 5 mm diameter pre-cleaned stainless steel punch, transferred to silver foil cups, treated with two 10 ml aliquots of 2 N HCl to remove carbonates [King *et al.*, 1998], dried at 60 °C, and analyzed using a Thermo-Finnigan Flash 1112 elemental analyzer and sulfanilamide standards at the University of Tasmania Central Science Laboratory. The >53  $\mu\text{m}$  fraction was treated in the same way, after first transferring the material from half the screen using pre-filtered (0.8  $\mu\text{m}$  GF/F) seawater onto 25 mm diameter 1.2  $\mu\text{m}$  pore size silver membrane filters (Sterlitech, Concord, MA, USA), and removing any large zooplankton manually with pre-cleaned forceps. Similarly, the PPS3/3 sediment trap samples were washed through a 350  $\mu\text{m}$  screen with this pre-filtered seawater (0.8  $\mu\text{m}$  GF/F filters) to remove zooplankton, and the particles also collected onto 25 mm silver membrane filters by low vacuum filtration. Blank corrections for the pump samples were estimated from pumps deployed in situ that failed to run, and for the trap samples by re-filtering the pre-filtered seawater. All blank corrections were less than 2% for all samples. The sub-sampling introduces uncertainties of 10-20% from inhomogeneous filter coverage that exceeds the analytical uncertainty of the POC analysis of ~1%, as discussed previously by Trull *et al.* [2008].

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