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Anna Dammshäuser, Thibaut Wagener, Peter L. Croot. Surface water dissolved aluminum and titanium: Tracers for specific time scales of dust deposition to the Atlantic?. *Geophysical Research Letters*, 2011, 38 (24), pp.n/a-n/a. 10.1029/2011gl049847 . hal-02433610

HAL Id: hal-02433610

<https://hal.science/hal-02433610>

Submitted on 12 May 2021

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Surface water dissolved aluminum and titanium: Tracers for specific time scales of dust deposition to the Atlantic?

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Received 29 September 2011; revised 8 November 2011; accepted 9 November 2011; published 17 December 2011.

[1] Surface water distributions of dissolved Al (dAl) and dissolved Ti (dT_i) were investigated along a meridional Atlantic transect and related to dust deposition estimates. In the zone of Saharan dust deposition, highest dAl concentrations occurred in the tropical salinity minimum and suggest increasing Al dissolution from Saharan aerosols with wet deposition. By contrast, the dT_i distribution is not related to precipitation but agrees with the pattern of annual dust deposition. In the zone of Patagonian dust deposition, elevated dT_i concentrations contrasted with decreased dAl concentrations, indicating excess dAl scavenging onto biogenic particles in surface waters. Estimated residence times range from months to years for dAl and are ~10 times higher for dT_i. This suggests that dAl reflects seasonal changes in dust deposition, while dT_i is related to longer temporal scales. However, spatial variations in input and removal processes complicate the quantification of dust deposition from surface water concentrations. **Citation:** Dammshäuser, A., T. Wagener, and P. L. Croot (2011), Surface water dissolved aluminum and titanium: Tracers for specific time scales of dust deposition to the Atlantic?, *Geophys. Res. Lett.*, 38, L24601, doi:10.1029/2011GL049847.

1. Introduction

[2] The deposition and dissolution of atmospheric lithogenic particles from continental sources (hereafter referred to as dust) is a major input path of trace elements into the open ocean. In recent years, the atmospheric supply of the potentially limiting nutrient iron has been a focus of intensive research [e.g., *Jickells et al.*, 2005], yielding a better understanding of dust dissolution processes and surface water residence times [e.g., *Baker and Croot*, 2010]. However, dust deposition to the open ocean is still poorly constrained and restricted to few direct observations [e.g., *Duce et al.*, 1991; *Luo et al.*, 2003, and references therein] and estimates based on model outputs [e.g., *Jickells et al.*, 2005; *Luo et al.*, 2003]. Dissolved Al (dAl) concentrations have been introduced as tracer to assess the supply of dust particles to the surface ocean [e.g., *Measures and Vink*, 2000]. By contrast, very little is known about the potential application of dissolved Ti (dT_i) concentrations to trace dust

input. The utilization of a certain element as a tracer for dust supply requires some estimations of its input, depending on the dust composition and solubility in surface seawater, and of its removal from the surface ocean. Surface water residence times constrain the time frame of dust deposition that can be inferred from the trace metal concentrations. Both Al and Ti are abundant, relatively invariant components of the earth's crust [*McLennan*, 2001] and are predominantly delivered to the open ocean by dust deposition [*Orians and Bruland*, 1986; *Orians et al.*, 1990]. In seawater, dAl and dT_i have scavenging type distributions and so far are not known to have essential biological functions. However, both elements also behave specifically in their dissolution from dust and have different residence times in the surface ocean [e.g., *Buck et al.*, 2010; *Orians and Bruland*, 1986; *Orians et al.*, 1990]. Therefore, Al and Ti may provide complementary approaches to assess the dust deposition to the surface ocean.

[3] In this study, the distribution of dAl and dT_i is investigated on a meridional Atlantic transect through ocean regions covering a wide range of dust deposition fluxes. The aim of this work was to identify controls on the relationship between surface water concentrations of dAl and dT_i and dust deposition and thus to better constrain the potential utilization of dAl and dT_i to trace dust inputs to the surface ocean.

2. Methods

[4] Surface seawater samples (20–25 m depth) were collected aboard the RV Polarstern (ANTXXVI-4, Punta Arenas–Bremerhaven) during April and May 2010. Water samples were collected using trace metal clean GO-FLO samplers (General Oceanics) and filtered through 0.2 μm cartridge filters (Sartobran® P, Sartorius) under slight nitrogen overpressure (0.2 bar). All sample handling and processing was conducted in the IFM-GEOMAR clean container (ISO 5, class 100). The samples were acidified with sub-boiled quartz distilled HCl to pH <2 at least 24 h before analysis and analyzed directly on board. Dissolved Al concentrations were determined using the method described by *Hydes and Liss* [1976]. In short, the reagent lumogallion is added, the samples are buffered to pH 5 with ammonium-acetate and heated to 50°C for 3 h to improve the complex formation. The fluorescence was measured with a Hitachi FL 2700 Fluorescence Spectrophotometer (excitation wavelength 497 nm, emission wavelength 572 nm). The detection limit was 0.1–0.3 nM for dAl, the blank values ranged between 0.4–0.6 nM on different days of analysis. Analysis of the SAFe reference seawater S1 resulted in 1.70 ± 0.21 nM of dAl ($n = 14$, consensus value 1.74 ± 0.09 nM). Dissolved Ti concentrations were determined by catalytic cathodic

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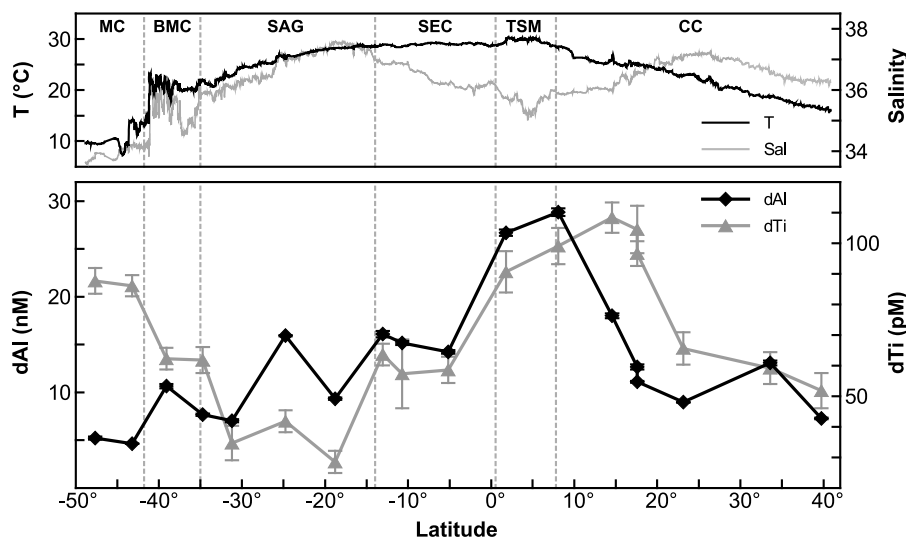


Figure 1. Surface seawater concentrations of dAl, dTi, salinity and temperature over the cruise transect. Dotted lines define the hydrographic regions encountered during the cruise (MC: Malvinas Current, BMC: Brazil Malvinas Confluence, SAG: South Atlantic Gyre, SEC: South Equatorial Current, TSM: Tropical Salinity Minimum, CC: Canary Current). Exact values and station locations are given in Table S1 in the auxiliary material.

stripping voltammetry (μ Autolab III with 663VA stand, Metrohm), using cupferron as the complexing ligand and bromate as an auxiliary oxidant [Croot, 2011]. The detection limit was 9 pM and the blank value 24 ± 3 pM for dTi. Supporting oceanographic data were obtained from continuous thermosalinograph measurements and the CTD system and are available in the Pangaea data management system [Rohardt, 2010; Rohardt and Bracher, 2011].

3. Results and Discussion

3.1. Surface Water Distributions of dAl and dTi

[5] Figure 1 displays dAl and dTi surface water concentrations over the cruise transect. The hydrographic regions were classified according to the variations in salinity and temperature [Tchernia, 1980]. Dissolved Al concentrations gradually increased from 5 nM in the Malvinas Current (MC) and the Canary Current (CC) towards 30 nM in the tropical salinity minimum (TSM), in agreement with previous observations [Measures et al., 2008; Vink and Measures, 2001]. Dissolved Ti concentrations showed two maxima, with concentrations of up to 90 pM in the MC and 110 pM in the CC at 15°N. Lowest concentrations of dTi (~ 30 pM) occurred in the South Atlantic Gyre (SAG). Few other studies report surface water concentrations of dTi in the Atlantic Ocean. Our results lay in the same range as earlier observations in the western North Atlantic [Orlans et al., 1990; Skrabal, 2006].

[6] In the tropical and subtropical North Atlantic, elevated concentrations of dAl and dTi agree with increased dust deposition [e.g., Duce et al., 1991; Jickells et al., 2005]. The additionally elevated concentrations of dAl at 33°N can be attributed to the propagation of Al-rich subtropical mode water from the western Atlantic basin [Measures et al., 2008]. Maximum concentrations of dTi occurred north of the highest dAl concentrations and are more in line with the location of highest annual dust deposition [Jickells et al., 2005]. In the South Atlantic, elevated dTi concentrations

indicate the input of dust from Patagonian sources [Johnson et al., 2010]. However, in this region dAl concentrations were decreased, with exception of elevated concentrations in the confluence of the Brazil and the Malvinas Currents (BMC) that can be attributed to the delivery of Al-rich equatorial waters with the Brazil Current (BC) [Vink and Measures, 2001]. Overall, our data suggest that specific input and/or removal processes determine the distribution of dAl and dTi in the different regions.

3.2. Residence Times of dAl and dTi

[7] In order to better assess the specific processes influencing dAl and dTi distributions, elemental residence times were calculated at each sampling station. The objective is to determine differences in the residence times of dAl and dTi rather than absolute values. For this calculation it was assumed that input solely occurs via dust deposition and that input and removal processes are balanced in the mixed layer. The residence time is thus given as quotient between soluble atmospheric flux and inventory of the dissolved metal. The inventory was estimated as integrated dissolved metal concentration over a mixed layer of 30 m (all samples originated from the mixed layer; see auxiliary material).¹ The atmospheric flux was estimated from the average annual dust deposition given in the model composite by Jickells et al. [2005, and references therein], assuming that over the Atlantic transect this estimate adequately reflects the spatial and temporal variations in dust deposition. The soluble flux was calculated assuming dust fractions of 8.04% and 0.41% for Al and Ti [McLennan, 2001] and solubilities of 5% and 0.1% respectively. The elemental fractions in the dust are well established [e.g., Formenti et al., 2003; Gaiero et al., 2007], however, the solubilities are key uncertainties in the residence time calculation. For Al, the solubility estimate is based on average values

¹Auxiliary materials are available in the HTML. doi:10.1029/2011GL049847.

Table 1. Residence Times of dAl and dTi Calculated From the Annual Average Dust Deposition Estimates by *Jickells et al.* [2005]^a

Region	Residence Time (years)		Residence time
	Al	Ti	Ti:Al
Malvinas Current	0.4–0.6	12–17	29–32
Brazil Malvinas Confluence	1.4–2.9	14–40	10–14
South Atlantic Gyre	4.6–17.2	39–78	5–9
South Equatorial Current	3.5–15.7	25–108	7
Tropical Salinity Minimum	0.7–1.8	4–10	6
Canary Current	0.2–1.1	3–14	5–18

^aFor details see section 3.2.

reported for the Atlantic [*Baker et al.*, 2006; *Prospero et al.*, 1987]. For Ti, an intermediate value was chosen based on results in seawater incubations with Saharan dust (auxiliary material) and aerosol filter leachings with deionized water [*Buck et al.*, 2010]. The observed variations in the Ti solubilities may reflect higher Ti solubility in deionized water, specific properties of aerosol and dust samples, or the experimental setup.

[8] In the regions of high dust deposition, the estimated residence times vary between months for dAl and several years for dTi (Table 1). Longer residence times occurred in the SAG, most likely due to decreased biological activity and slower removal by scavenging processes. For Al, the observed ranges agree with previous estimations, in the Sargasso Sea [*Jickells*, 1999], the North Pacific [*Orians and Bruland*, 1986] and from modeling approaches [*Gehlen et al.*, 2003; *Han et al.*, 2008]. For dTi, to our knowledge these are the first residence time estimates in Atlantic surface waters. The residence times of dTi clearly exceed those of dAl, on average by a factor of around 10. This generally indicates higher reactivity of dAl in the surface ocean. Specific differences occurred in the MC and the TSM, and could be related to spatial variations in both inventories and/or atmospheric fluxes of dAl and dTi in these regions.

3.3. Relationship Between dAl and dTi Surface Water Concentrations and Dust Deposition

[9] Surface water dAl concentrations have been previously suggested as tracer to estimate the dust supply to the ocean, giving reasonable agreement to model based estimates of dust deposition [*Han et al.*, 2008; *Measures and Vink*, 2000]. Our observed dAl concentrations similarly agree with the deposition of Saharan dust in the tropical North Atlantic. However, the dAl concentrations do not reflect the deposition of dust from Patagonian sources that is suggested in a more recent estimate [*Jickells et al.*, 2005], whereas dTi concentrations were elevated in both regions. Furthermore, dAl and dTi were specifically distributed within the Saharan dust input region. These observations are in contrast to the expected common atmospheric supply of both metals. Potentially influencing factors for the different regions are discussed below.

3.3.1. Patagonian Dust Input Region

[10] In the MC, decreased dAl concentrations contrast with increased dTi concentrations, and the difference between dAl and dTi residence times is more pronounced than in other regions (Figure 1 and Table 2). The differences

could result either from additional supply of dTi, or increased removal of dAl in this region. The input of excess dTi from atmospheric sources is unlikely, since Ti/Al ratios from Patagonian and Saharan dust are similar [*Formenti et al.*, 2003; *Gaiero et al.*, 2007] and solubility experiments by *Baker et al.* [2006] rather indicate higher solubility of Al for Patagonian than for Saharan dust. The sampling stations in the MC were located at the edge of the Patagonian shelf (300–400 km offshore), suggesting that additional supply of dTi might have occurred via coastal erosion or resuspension of shelf sediments [*Gaiero et al.*, 2003; *Pierce and Siegel*, 1979]. Suspended material may be transported offshore with surface currents [*Pierce and Siegel*, 1979], however, previous studies showed only small release of dTi through sediment resuspension [*Skrabal and Terry*, 2002] compared to considerable release of dAl [*Moran and Moore*, 1991; *Skrabal and Terry*, 2002]. Similarly, pore water profiles indicate the benthic supply of both metals to bottom waters [*Skrabal and Terry*, 2002]. The supply of excess dTi over dAl in the MC is thus unlikely, suggesting that the deviating distributions and residence times of dAl and dTi rather result from the preferential removal of dAl over dTi. The removal of dAl is strongly influenced by surface adsorption on biogenic particles and/or biological uptake, and is thus coupled to biological productivity [e.g., *Gehlen et al.*, 2002; *Middag et al.*, 2009; *Moran and Moore*, 1988]. Observations in sediment cores indicate that dTi is less susceptible to such removal than dAl [e.g., *Murray and Leinen*, 1996]. SeaWiFS images show high chlorophyll a concentrations in the MC in the two months preceding the sampling (auxiliary material). This suggests that the removal with biogenic particles may indeed have resulted in excess removal of dAl over dTi. The high reactivity of dAl would also explain why dAl concentrations in April do not reflect the Patagonian dust deposition that occurs predominantly in austral summer [*Johnson et al.*, 2010].

3.3.2. Saharan Dust Input Region

[11] In the North Atlantic, the dAl maximum concurred with the TSM as well as with the southward shifted zone of maximum Saharan dust transport during boreal winter [*Moulin et al.*, 1997]. In agreement with short residence times, the dAl concentrations may thus reflect the seasonal dust deposition pattern in the months preceding the sampling. However, previous studies (Table 2) similarly found highest dAl concentrations in the TSM in boreal summer [*Measures et al.*, 2008], that do not agree with the seasonal

Table 2. Observations of Maximum Al Concentrations in the Tropical and Subtropical Atlantic

Study	Sampling period	Location of Maximum
<i>Total Dissolvable Al</i>		
<i>Helmers and Rutgers van der Loeff</i> [1993]	May	7°S–10°N
<i>Helmers and Rutgers van der Loeff</i> [1993]	Oct	2°S–20°N
<i>Bowie et al.</i> [2002]	May–Jun	8°S–10°N
<i>Bowie et al.</i> [2002]	Sep–Oct	10°N–25°N
<i>Dissolved Al</i>		
<i>Measures et al.</i> [2008]	Jul–Aug	0–10°N
This study	April–May	0–10°N

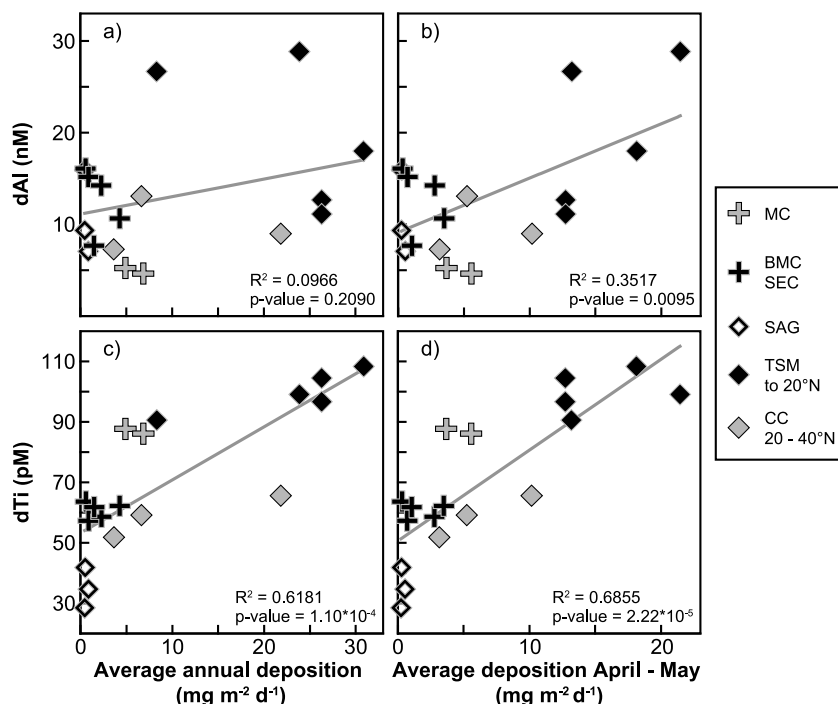


Figure 2. Dissolved Al and dTi concentrations versus average dust deposition estimates from the model composite by *Jickells et al.* [2005]. (a and c) show the average annual dust deposition and (b and d) the average dust deposition during the sampling period (April–May). The grey lines indicate the correlation functions. For further details refer to section 3.3.3.

dust deposition pattern. In contrast to this, the maximum concentrations of total dissolvable Al reflected the seasonal movement of the maximum dust transport between boreal spring and autumn (Table 2) [Bowie *et al.*, 2002; *Helmers and Rutgers van der Loeff*, 1993]. This suggests that, due to increased solubility of Al with the lower pH in rainwater [Losno *et al.*, 1993; Prospero *et al.*, 1987], dAl is predominantly supplied by wet deposition within the Inter-Tropical Convergence Zone. The underestimated solubility of Al in rainwater could have resulted in an overestimation of the residence times in the TSM. By contrast to dAl, the distribution of dTi in the North Atlantic agreed with the annual dust deposition average [Jickells *et al.*, 2005] and the estimated longer residence times for dTi. The absence of elevated dTi concentrations in the TSM suggests that potential solubility differences between rainwater and seawater are less pronounced for Ti than for Al. Overall, the different distribution of dAl and dTi in the North Atlantic can be explained by two processes that may also be complementary: a) due to different residence times the dAl and dTi concentrations reflect the variations between seasonal and annual dust deposition and b) the difference between the dust solubilities in rainwater and seawater is more pronounced for dAl, resulting in a stronger influence of wet deposition for dAl than for dTi.

3.3.3. Relationship Over the Atlantic Transect

[12] Figure 2 shows the relationship between the surface water concentrations of dAl and dTi and dust deposition estimates given in the model composite by *Jickells et al.* [2005, and references therein] for both the annual and the sampling period (April–May) average. For dAl the correlation improves if the seasonal dust deposition is considered (Figures 2a and 2b), suggesting that dAl concentrations are

stronger related to integrations of the dust deposition over shorter time scales. By contrast, dTi concentrations are reasonably correlated with both the annual and the seasonal dust deposition estimate (Figures 2c and 2d). These observations support the idea that dAl concentrations reflect seasonal variations in the dust deposition, whereas dTi concentrations rather reflect interannual variations in the dust deposition. This suggests that dAl and dTi concentrations may serve as tracers for dust deposition variations over different time scales. However, in addition to the general relationship, dAl and dTi distributions show specific spatial variations over the Atlantic transect (see data points from MC in Figure 2). As discussed above, additional differences between both metals can be attributed to the excess removal of dAl in the MC and to the elevated supply of only dAl with wet deposition in the TSM. Previous studies suggested the simple estimation of dust inputs from dAl concentrations with the assumption of invariant values for both residence times (5 years) and soluble dust fractions of Al (1.5–5%) [e.g., *Measures and Vink*, 2000; *Measures et al.*, 2008; *Vink and Measures*, 2001]. However, our data indicate variations in both factors in different regions of the Atlantic Ocean. Considering the observed residence times (Table 1), such calculation would likely overestimate dust inputs in the SAG (dAl residence times >5 years) and underestimate inputs in the MC, TSM and CC (dAl residence times <5 years). Similarly, underestimated solubilities of dust Al in regions with strong influence of wet deposition would result in overestimated dust inputs.

4. Conclusion

[13] Despite the common source of dAl and dTi we observed distinct differences in the distributions of both

metals in Atlantic surface waters. Patagonian dust input was only reflected in dTi concentrations, most likely due to fast removal of dAl onto biogenic particles. In the North Atlantic, highest dAl concentrations indicate the increased dissolution and supply of dust derived Al with wet deposition. By contrast, the distribution of dTi was not related to wet deposition but reflected the pattern of the average annual dust deposition [Jickells *et al.*, 2005]. Residence time estimates and the relation between surface water concentrations and dust deposition suggest that dAl concentrations may serve to trace seasonal variations in the dust deposition, whereas dTi concentrations may serve to trace variations in the dust deposition over longer time scales. However, our data also indicate spatial differences in supply and removal processes of dAl and dTi in the surface ocean. Variations in solubilities and residence times have to be considered if the dust deposition is assessed from surface water concentrations of dAl and dTi.

[14] **Acknowledgments.** We thank the officers and crew of RV Polarstern and our colleagues D. Gaiero and M. Heller for their help during ANTXXVI-4, and two anonymous reviewers for their constructive comments on the manuscript. Funding for the participation and support of the work performed during ANTXXVI-4 was provided by the DFG via grants to PLC (CR145/15-1 and CR145/18-1). The financial support by SOPRAN (BMBF FKZ 03F0462A and 03F0611A) is also gratefully acknowledged. T. Wagener was supported by a Marie Curie IEF (grant agreement PIEF-GA-2009-236694, DAPOP).

[15] The Editor wishes thank two anonymous reviewers for their assistance evaluating this paper.

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