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Estimation of atmospheric nutrient inputs to the Atlantic Ocean from 50°N to 50°S based on large-scale field sampling: Fixed nitrogen and dry deposition of phosphorus

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[1] Atmospheric nitrogen inputs to the ocean are estimated to have increased by up to a factor of three as a result of increased anthropogenic emissions over the last 150 years, with further increases expected in the short- to mid-term at least. Such estimates are largely based on emissions and atmospheric transport modeling, because, apart from a few island sites, there is very little observational data available for atmospheric nitrogen concentrations over the remote ocean. Here we use samples of rainwater and aerosol we obtained during 12 long-transect cruises across the Atlantic Ocean between 50°N and 50°S as the basis for a climatological estimate of nitrogen inputs to the basin. The climatology is for the 5 years 2001–2005, during which almost all of the cruises took place, and includes dry and wet deposition of nitrate and ammonium explicitly, together with a more uncertain estimate of soluble organic nitrogen deposition. Our results indicate that nitrogen inputs into the region were ~850–1420 Gmol (12–20 Tg) N yr^{-1}, with ~78–85% of this in the form of wet deposition. Inputs were greater in the Northern Hemisphere and in wet regions, and wet regions had a greater proportion of input via wet deposition. The largest uncertainty in our estimate of dry inputs is associated with variability in deposition velocities, while the largest uncertainty in our wet nitrogen input estimate is due to the limited amount and uneven geographic distribution of observational data. We also estimate a lower limit of dry deposition of phosphate to be ~0.19 Gmol P yr^{-1}, using data from the same cruises. We compare our results to several recent estimates of N and P deposition to the Atlantic and discuss the likely sources of uncertainty, such as the potential seasonal bias introduced by our sampling, on our climatology.

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1. Introduction

[2] The global fixed nitrogen cycle (i.e., that excluding relatively inert N_2 gas) has been dramatically altered by human activity, particularly over the last 150 years [e.g., *Galloway et al.*, 2008]. This perturbation has resulted in greatly increased atmospheric transport of fixed nitrogen to the oceans which may be enhancing ocean productivity, CO_2 uptake and N_2O emissions [*Duce et al.*, 2008]. Current best estimates of nitrogen inputs to the ocean (Table 1) emphasize the significance of the atmospheric flux, but it is apparent that the uncertainty in these fluxes is also large. The atmospheric

fluxes in Table 1 are based on the averaged results of several models (*Dentener et al.* [2006], as modified by *Duce et al.* [2008]) and, while this average is in good agreement with (land-based) measurements, it is clear from the between-model variability in that study that substantial uncertainties in the global atmospheric nitrogen cycle still remain. The atmospheric nitrogen cycle over the oceans must be considered the most uncertain part of such global modeling because the validation of model output is primarily based on comparisons to terrestrial sampling sites, as the global network of long-term monitoring sites is overwhelmingly terrestrial [*Dentener et al.*, 2006]. The impact of atmospheric nitrogen deposition varies regionally, with highest impacts occurring where atmospheric inputs are large and water column nitrogen concentrations are low [*Duce et al.*, 2008].

[3] The atmospheric nitrogen cycles over land and the sea differ significantly, due for instance to air-sea ammonia exchange [*Johnson et al.*, 2008] and the interactions between sea spray and nitric acid [*Jickells*, 2006], which acts to move nitrate from fine to coarse mode aerosol in the marine

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Table 1. Current Fixed Nitrogen Inputs to the World Oceans[a]

Source	Flux (10^{12} mol yr^{-1})
Biological N_2 fixation	7.1 (4.2–14.3)
Rivers	4.7 (3.6–5.7)
Atmospheric	4.8 (2.7–6.8)

[a]From *Duce et al.* [2008].

atmosphere, and thereby substantially increases its dry deposition rate. The major pools of soluble nitrogen in aerosol and rainwater are nitrate, ammonium and organic nitrogen. Nitrate is predominantly derived from nitrogen oxides (NO_x) emitted through combustion processes, while the dominant source of ammonium is emissions of ammonia associated with agricultural activity [*Aneja et al.*, 2001]. The origins of the organic matter are much less certain [*Cornell et al.*, 2003] and the distribution of this component and its possible origins are discussed elsewhere [*Zhang et al.*, 2008; *Lesworth et al.*, 2010]. *Russell et al.* [2003] have noted the possible existence of additional organic nitrogen in aerosols that is not water soluble. We do not discuss the air-sea exchange of ammonia here. It is not currently clear if the aerosol samples collected will include ammonia gas as well as ammonium aerosol. Both components can be significant in some parts of the marine atmosphere although the gas phase species is short-lived with respect to removal to aerosol or seawater [*Norman and Leck*, 2005]. However, the overall air-sea flux of ammonia is estimated to be very small (0 ± 140 Gmol N yr^{-1} globally (M. T. Johnson et al., manuscript in preparation, 2010)) compared to the total air-to-sea fixed nitrogen flux considered here, and hence does not significantly influence the overall fluxes, although the ammonia flux is important to aerosol formation and atmospheric pH [*Zhuang and Huebert*, 1996; *Keene et al.*, 1998].

[4] Phosphorus in the atmosphere is predominantly associated with the particle phase and is derived from mineral dust, biomass and fossil fuel combustion, seaspray and volcanic ash. *Mahowald et al.* [2008] have recently discussed the relative importance of these sources and have made the first attempt to model the global distribution and deposition flux of atmospheric phosphorus.

[5] The discussion above highlights the importance of assessing atmospheric nutrient supply to the oceans using field sampling in addition to model results. However, our ability to do this is severely limited by the lack of island sampling sites which allow long-term monitoring of deposition over the oceans, and by the restricted geographical location of those sites for which data have been obtained [e.g., *Dentener et al.*, 2006]. In this work we attempt to derive atmospheric nitrogen inputs to the Atlantic Ocean using data obtained from aerosol and rain sampling on cruises that traverse large areas of the Atlantic Ocean over a six year period. While the results of a few of these cruises have been published elsewhere [*Baker et al.*, 2003, 2006a, 2007], the goal of this paper is to draw all our data together and estimate nitrogen inputs to surface Atlantic waters between 50°N and 50°S. We have previously observed that nutrient concentrations in aerosol samples collected over the Atlantic are heavily influenced by the source regions that the air sampled has

passed over before collection [*Baker et al.*, 2006a]. Here we extend the database reported in that work, to include a total of 12 research cruises covering a wider region of the Atlantic, providing a uniquely large data set from which to estimate atmospheric nutrient inputs. We take advantage of the source region dependency of atmospheric nitrogen concentrations to derive a climatology of atmospheric dry nitrogen fluxes to the basin. This dry flux estimate is combined with a climatology of wet nitrogen deposition flux derived from our field measurements and we discuss the relative contributions of the two deposition modes and the spatial distribution of atmospheric nitrogen inputs to the Atlantic. Since current models of the global atmospheric nitrogen cycle generally consider nitrate and ammonium, but not organic nitrogen explicitly, we focus on these two former species, but we also consider the potential contribution of soluble organic nitrogen to deposition to the basin. We also attempt to estimate the dry deposition of soluble phosphate to the Atlantic, using data from the same cruises, and examine the relative input rates of nitrogen and phosphate from the atmosphere, since these represent the two primary limiting nutrients over much of the world ocean [*Moore et al.*, 2002]. Elsewhere we use a similar data set to address the atmospheric input of iron, the other key limiting nutrient, and other elements associated with mineral dust, to the Atlantic (A. R. Baker et al., manuscript in preparation, 2010).

2. Methods

2.1. Sampling and Analysis

[6] Table 2 lists the dates and start/end points of the cruises during which samples have been collected, and the cruise tracks and aerosol sampling start locations are shown in Figure 1. Sampling methods have been described elsewhere [*Baker et al.*, 2006a, 2007] and for aerosols generally consisted of collection over approximately 24 h with the ship sailing into the wind to avoid ship-based contamination. High volume aerosol sampling ($\sim 1 m^3$ min^{-1}) onto Whatman 41 filters was used and samples were either collected using Sierra-type cascade impactors with separation into coarse ($>1\mu m$ diameter) and fine ($<1\mu m$ diameter) particles, or using a single bulk Whatman 41 filter (see Table 2). Whatman 41 papers are known to also collect gas phase nitric acid, but concentrations of nitric acid in the remote marine atmosphere are low and the contribution to nitrate concentrations should therefore be low [*Baker et al.*, 2003]. Filters were generally used without any pretreatment, although for AMT16 and ANT23-1 they were washed with dilute hydrochloric acid before use in order to reduce trace metal blanks. Acid washing may have induced some passive uptake of ammonia on these filters, increasing their ammonium blanks, but the effect was not large (see Table S1).[1] For each cruise, blanks for nitrogen species were assessed by analyzing unexposed filters and filters that had been deployed in the cascade impactor or filter cassette alone, or in the cascade impactor or filter cassette that was then deployed in the collector

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[1]Auxiliary materials are available with the HTML. doi:10.1029/2009GB003634.
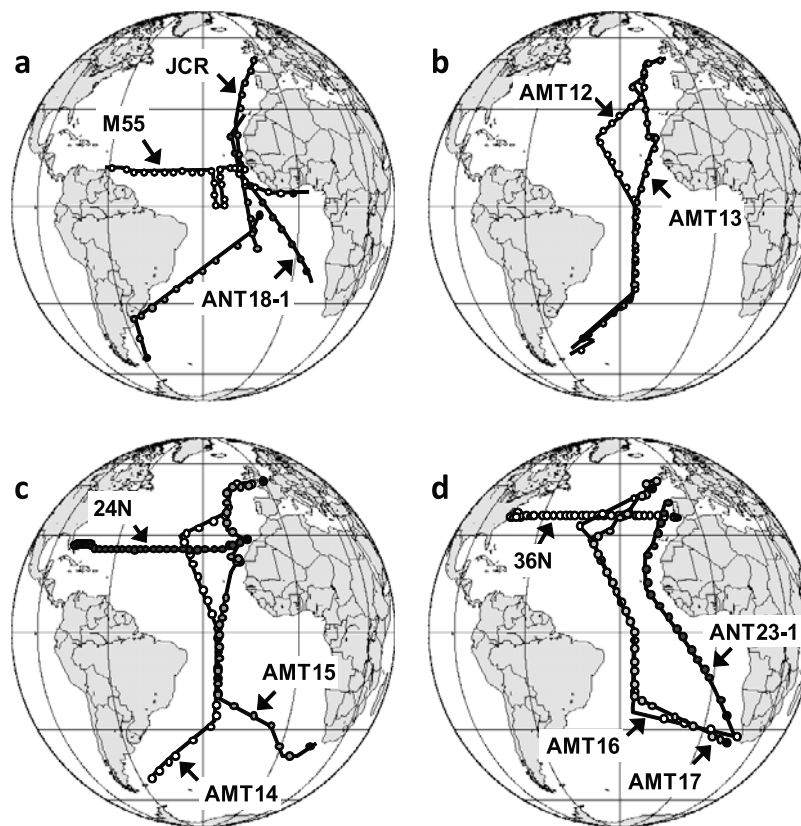
Figure 1. Aerosol sampling locations used in this work for cruises which took place in (a) 2000–2002, (b) 2003, (c) 2004, and (d) 2005. Markers show sampling start positions, with aerosol collection generally continuing until the beginning of the following sample. Sampling end positions for each cruise are shown by black markers.

without the motor running. Results of these blanks determinations are reported in Table S1.

[7] Rain samples were collected by manually opening rain collectors (low density polyethylene bottles attached to 28 cm or 40 cm diameter polypropylene funnels) immediately prior to, or at the onset of, rain and closing them upon the cessation of rain. Rain sampling equipment was cleaned by soaking in detergent (10% v/v Decon 90) and then deionized water for at least 48 h each before use, as described by *Baker et al.* [2007]. Blanks for rain sampling were determined during each cruise by collecting ultrapure water that had been used to rinse the collection surfaces of the rain funnels and determining the nitrogen content of

Table 2. Details of Cruises From Which Data Have Been Used in This Study[a]

Cruise	Dates	Start	End	N_a (Type)	N_r
ANT18-1[b,c]	29 Sept to 23 Oct 2000	Bremerhaven, Germany	Cape Town, South Africa	9 (b)	1
JCR[b,c]	10 Sept to 24 Oct 2001	Grimsby, United Kingdom	Port Stanley, Falkland Islands	25 (ss)	6
M55[d]	12 Oct to 17 Nov 2002	Curacao, Caribbean	Douala, Cameroon	27 (ss)	16
AMT12[c]	12 May to 17 Jun 2003	Port Stanley, Falkland Islands	Grimsby, United Kingdom	23 (ss)	7
AMT13[c]	10 Sept to 14 Oct 2003	Immingham, United Kingdom	Port Stanley, Falkland Islands	22 (ss)	11
24N	4 Apr to 10 May 2004	Freeport, Bahamas	Tenerife, Canary Islands	32 (b)	–
AMT14	28 Apr to 1 Jun 2004	Port Stanley, Falkland Islands	Grimsby, United Kingdom	26 (ss)	13
AMT15	17 Sept to 28 Oct 2004	Southampton, United Kingdom	Cape Town, South Africa	26 (ss)	10
36N	1 May to 15 Jun 2005	St. Georges, Bermuda	Lisbon, Portugal	33 (ss)	6
AMT16	20 May to 29 Jun 2005	Cape Town, South Africa	Falmouth, United Kingdom	14 (b)	–
ANT23-1	13 Oct to 17 Nov 2005	Bremerhaven, Germany	Cape Town, South Africa	17 (b)	–
AMT17	15 Oct to 28 Nov 2005	Glasgow, United Kingdom	Port Elisabeth, South Africa	28 (ss)	13

[a]Including number of aerosol (N_a) and rain (N_r) samples and type of aerosol samples (b, bulk; ss, size segregated).
[b]*Baker et al.* [2003].
[c]*Baker et al.* [2006a].
[d]*Baker et al.* [2007].

Figure 2. Locations of rainfall samples collected during this work. Symbol shapes indicate cruises which took place in 2000–2002 (squares), 2003 (circles), 2004 (diamonds), and 2005 (triangles), with symbol shadings corresponding to those used for aerosol sampling locations in Figure 1. Tracks of cruises during which rainfall was not collected are indicated by dashed lines.

this rinse water (see Table S2). The locations of rain samples are shown in Figure 2.

[8] Samples were stored frozen and later thawed and analyzed using methods discussed elsewhere [*Baker et al.*, 2006a, 2007; *Lesworth et al.*, 2010] which for the components of interest here involves ion chromatographic analysis for nitrate and ammonium and high temperature catalytic oxidation for total soluble nitrogen (TSN) on water extracts of filters and filtered rain samples. Soluble organic nitrogen (SON) was estimated as the difference between total soluble nitrogen and nitrate plus ammonium and the sum of errors arising from this calculation makes the estimations of soluble organic nitrogen relatively more uncertain than nitrate and ammonium [*Cornell et al.*, 2003]. Data quality for SON and the estimation of SON inputs to the Atlantic are discussed in Section 3.4. Soluble phosphate was determined using spectrophotometry in aqueous extracts (pH 7) of aerosol samples [*Baker et al.*, 2006a, 2007].

2.2. Climatology and Air Mass Back Trajectories

[9] Because our database for rainfall samples (n = 81) is smaller than for aerosols (n = 283) and is geographically biased to certain regions, consistent with the known distribution of rainfall over the Atlantic [*Xie and Arkin*, 1997], we have based our wet deposition estimate on input to 5 broad regions of the Atlantic, corresponding to areas of relatively high and low precipitation. These are the North Atlantic storm track (R1) and dry region (R2), the Inter-tropical Convergence Zone (ITCZ, R3) and the South Atlantic dry region (R4) and storm track (R5) (Figure 3). Approximately half of our rain samples were collected in the ITCZ, and we have therefore

subdivided that region at the 25°W meridian into western (R3a) and eastern (R3b) portions.

[10] We take advantage of the larger database for aerosol samples to further subdivide the 5 broad, wet deposition, regions in order to estimate dry deposition fluxes (Figure 3). The distribution of these sub-regions reflects the spatial density of data in our database, so that we have more (smaller) boxes in the North Atlantic than in the South Atlantic. This provides a higher resolution dry deposition estimate, which can be aggregated with the coarser wet deposition grid. Our approach in compiling the dry deposition climatology is based on the observation that aerosol concentrations over the Atlantic (as elsewhere) are strongly dependent on air mass origin and transport [e.g., *Baker et al.*, 2006a]. We therefore use air mass back trajectory classification as the basis for our dry deposition estimate. For individual aerosol samples collected during cruises we obtained 5-day air mass back trajectories for heights of 10, 500 and 1000m above the ships' positions from the NOAA HYSPLIT model (FNL data set). Each cruise sample was then assigned to one of 8 air mass types, according to which of the major aerosol source regions the trajectories had passed over within that 5 day period. Similar to *Baker et al.* [2006a], most
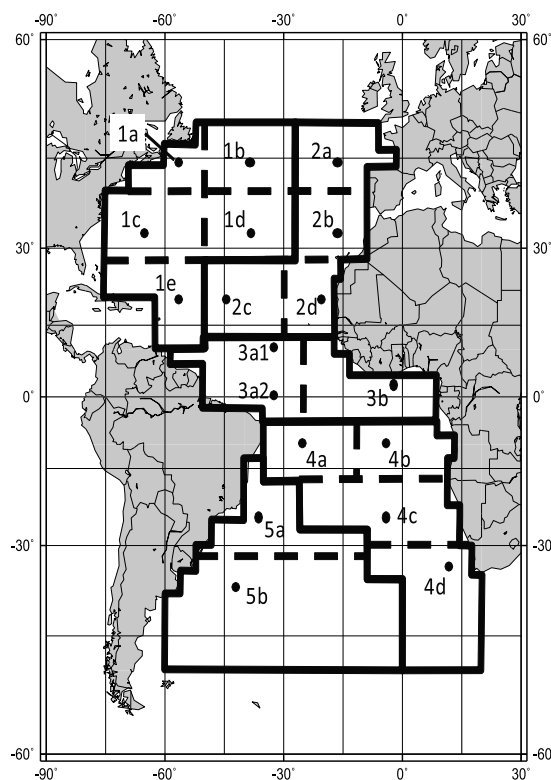
Figure 3. Map showing the boundaries of the wet (solid lines) and dry (dashed lines) deposition regions into which the Atlantic was divided for this study. Points (1a–5b) at which air mass back trajectories were obtained for air mass climatology determination in each box are also shown and this numbering also identifies the dry deposition regions.

Table 3. Median Fractions (f_c) of Aerosol Nitrate, Ammonium, and Phosphate Present on Coarse-Mode Particles for Each Air Mass Type Sampled During This Work[a]

Air Mass	$f_c(NO_3)$	$f_c(NH_4)$	$f_c(PO_4)$
NAmer	0.83 (10)	0.16 (10)	0.49 (10)
NAtl-Rem	0.91 (58)	0.18 (58)	0.53 (43)
Eur	0.90 (9)	0.13 (9)	0.43 (9)
Sahara	0.91 (55)	0.09 (55)	0.48 (48)
SAfr	0.91 (22)	0.13 (22)	0.53 (21)
SAfr-BB	0.95 (16)	0.13 (16)	0.49 (13)
SAtl-Rem	0.87 (38)	0.23 (34)	0.40 (19)
SAmer	0.77 (6)	0.20 (6)	0.44 (6)

[a]Number of samples for each case are given in parentheses.

of the air mass types correspond to the major continental landmasses surrounding the basin: North America (NAmer), Europe (Eur), North Africa (Sahara), Southern Africa (SAfr) and South America (SAmer). We also assigned air mass types to the cases where remote air had been circulating over the ocean (in either hemisphere) for the 5 days prior to collection (NAtl-Rem and SAtl-Rem) and to the case during austral spring where biomass burning emissions from Southern Africa were encountered (SAfr-BB). The only exception to this was for samples collected in the (sub-)tropical North Atlantic, which exhibited the characteristic yellow/orange/brown coloration typical of mineral dust. Such samples were classified as Saharan, regardless of whether their 5-day air mass back trajectories reached North Africa. These same air mass types were then used as the basis for compiling an air mass climatology for the Atlantic which is used to weight the dry deposition contributions from the various sources (see below). In this case, 5-day air mass back trajectories were obtained for each day of a 5 year period (2001-5) for a number of fixed points distributed through the basin (see Table A1 and Figure 3). These were obtained at starting heights of 50, 500 and 1000m from the British Atmospheric Data Centre (BADC) based on the ECMWF archive from the online web server (http://badc.nerc.ac.uk/community/trajectory/). A full description of the climatology is given in Appendix A, and example trajectories for each air mass type are included in Figures S1 and S2.

2.3. Flux Calculations

[11] Wet deposition fluxes (F^w) were estimated according to equation 1, in which C_R represents the concentration of each species in rainwater (see Section 3.1) and P is the precipitation rate. The latter was calculated from the 2.5x2.5° gridded monthly output of the CMAP model updated from *Xie and Arkin* [1997] (http://www.cdc.noaa.gov/cdc/data.cmap.html).

$$F^w = C_R P \qquad (1)$$

[12] Dry deposition fluxes (F^d) are strongly dependent on aerosol particle size and wind speed. In this analysis F^d is calculated as the product of the surface layer aerosol concentration and deposition velocity (v_d). The latter is calculated based on a modified parameterization for particle dry

deposition to "smooth" natural waters by *Slinn and Slinn* [1980]. It accounts for the enhancement of dry deposition by whitecaps [*Hummelshøj et al.*, 1992] and the effect of particle growth for a large relative humidity, increasing the effective dry deposition velocity by an order of magnitude compared to dry particles [*Ganzeveld et al.*, 1998]. We calculated dry deposition velocities for each sub-region using climatological mean wind speeds for the period 2001–2005 obtained from the ECMWF ERA-Interim data set and chose particle sizes of 0.1 μm for submicron particles and 7 μm for supermicron particles, with the latter being derived from the observed distribution of nitrate in Atlantic aerosols reported by *Keene et al.* [2009]. The calculated deposition velocities fall in the range 0.02–0.03 and 0.6–1.2 cm s^{-1} for fine and coarse modes respectively. Aerosol concentrations (C_A) used in the dry flux calculation (equation 2) were the median values for fine (f) and coarse (c) fractions of the relevant subset of our database (see below). Uncertainties in deposition velocities are discussed in Section 4.2. For some of the cruises we report here (~25% of the total sample numbers) we do not have aerosol size distribution data (see Table 2). In those cases we calculated the concentrations of each species in the fine and coarse modes based on the median fine/coarse size distribution of each species for each air mass type, as determined from the rest of our database (Table 3), and used these calculated fine and coarse concentrations in the dry flux calculation.

$$F^d = C_A^c v_d^c + C_A^f v_d^f \qquad (2)$$

3. Results

3.1. Wet Deposition: Rainfall Chemical Characteristics and Deposition Fluxes

[13] In Table 4 we show range and median concentration values for nitrate and ammonium determined in our rainwater samples for each of the wet deposition regions, and for the data set as a whole. Our results are broadly similar to those of previous studies within our study region, although most published data for rainwater nitrogen concentrations relate to samples collected in our Region 1. For example, nitrate concentrations in the range 0–19.4 μmol L^{-1} and ammonium concentrations in the range 0–8.9 μmol L^{-1} were reported for samples collected between the east coast of North America and Bermuda [*Galloway et al.*, 1983] and *Losno et al.* [1991] measured 19 μmol L^{-1} nitrate in one sample from the east of our Region 1. *Willey and Cahoon* [1991] reported a volume-weighted-mean (VWM) nitrate concentration of 8.6 μmol L^{-1} for 10 samples collected off the coast of North and South Carolina. Long-term monitoring of precipitation at Bermuda produced VWM concentrations for nitrate and ammonium of ~4.6 μmol L^{-1} and ~3 μmol L^{-1} respectively (J. N. Galloway, personal communication, 2009), while VWM concentrations at Barbados were ~2.8 μmol L^{-1} for both nitrate and ammonium (J. N. Galloway, personal communication, 2009). Outside Region 1 we have identified only the results of *Losno et al.* [1991], who reported nitrate concentrations of 3 μmol L^{-1} (n = 1) in

Table 4. Range and (in parentheses) Median Concentrations and Number of Samples for Nitrate and Ammonium and Their Volume-Weighted-Mean Concentrations (C_R) for the Rain Samples Collected in Each of the Wet Deposition Regions and for All the Rainfall Samples

Region	NO_3 (μmol L^{-1})	C_R^{NO3} (μmol L^{-1})	NH_4 (μmol L^{-1})	C_R^{NH4} (μmol L^{-1})
1, NAtl. Storm	1.4–62.9 (10.1, 14)	8.1	<0.5–14.8 (1.9, 14)	2.6
2, NAtl. Dry	2.8–33.7 (5.9, 7)	7.8	<0.4–15.0 (2.2, 6)	7.8
3a, ITCZ (west)	1.2–15.4 (3.3, 22)	3.5	0.3–7.3 (1.5, 22)	2.0
3b, ITCZ (east)	0.8–38.9 (5.6, 19)	5.4	<0.1–68.4 (6.9, 18)	7.9
4, SAtl. Dry	<0.2–22.8 (4.8, 6)	4.5	<0.4–9.4 (1.2, 6)	1.4
5, SAtl. Storm	<1.6–72.0 (5.9, 13)	6.7	<0.8–43.8 (3.9, 13)	6.6
All	<0.2–72.0 (5.0, 81)	5.3	<0.1–68.4 (2.0, 79)	5.2

Region 2, < 2–25 μmol L^{-1} (n = 4) in Region 3a and 2–38 μmol L^{-1} (n = 5) in Region 5.

[14] Concentrations in rainfall are strongly dependent on rainfall amount, in addition to source region, reflecting the well known dilution effect [*Jaffrezo et al.*, 1990]. We therefore calculated VWM rainfall concentrations (C_R) from the concentrations (C_i) and volumes (V_i) for all the rain samples collected (equation 3) within each of the 5 deposition regions (Table 4). In some cases, one or other of the nitrogen species was present at concentrations below our limits of detection. When this occurred we substituted a value equal to 75% of

the limit of detection for that species' concentration in the calculation of C_R.

$$C_R = \frac{\sum C_i V_i}{\sum V_i} \tag{3}$$

[15] The wet deposition flux of each N species in each deposition region was then calculated from C_R and the median precipitation in that region [after *Xie and Arkin*, 1997] using equation 1. Using this approach we estimate that wet deposition contributes 330 Gmol N yr^{-1}

Table 5. Wet and Dry Deposition Fluxes and Total Annual Inputs of Nitrate to the Five Broad Deposition Regions

Region	Sub-region	Wet Flux (μmol m^{-2} d^{-1})	Wet Input (Gmol yr^{-1})	Dry Flux (μmol m^{-2} d^{-1})	Dry Input (Gmol yr^{-1})	Total Flux (μmol m^{-2} d^{-1})	Total Input (Gmol yr^{-1})
1	1a			15.9	5.8		
	1b			13.2	9.7		
	1c			13.6	15.7		
	1d			7.4	7.8		
	1e			5.4	7.0		
Total		24.1	111	10.0	46.0	34.1	157
2	2a			11.0	7.5		
	2b			10.3	8.8		
	2c			7.5	9.6		
	2d			16.4	14.2		
Total		10.1	37.1	10.9	40.1	21.0	77.2
3	3a	16.8	32.3	5.4	10.3	22.2	42.6
	3b	19.2	34.9	4.4	8.1	23.6	43.0
Total		18.0	67.2	4.9	18.4	22.9	85.6
4	4a			6.7	8.5		
	4b			2.4	3.1		
	4c			2.6	4.7		
	4d			2.5	3.9		
Total		1.9	11.2	3.4	20.2	5.3	31.3
5	5a			1.9	2.9		
	5b			2.5	8.6		
Total		20.9	103	2.3	11.5	23.3	115
All		14.4	330	5.9	136	20.4	466

Table 6. Wet and Dry Deposition Fluxes and Total Annual Inputs of Ammonium to the Five Broad Deposition Regions

Region	Sub-region	Wet Flux (μmol m^{-2} d^{-1})	Wet Input (Gmol yr^{-1})	Dry Flux (μmol m^{-2} d^{-1})	Dry Input (Gmol yr^{-1})	Total Flux (μmol m^{-2} d^{-1})	Total Input (Gmol yr^{-1})
1	1a			3.9	1.4		
	1b			3.4	2.5		
	1c			3.4	3.9		
	1d			2.0	2.1		
	1e			1.6	2.1		
Total		7.7	35.3	2.6	12.0	10.3	47.3
2	2a			1.6	1.1		
	2b			1.3	1.1		
	2c			1.0	1.3		
	2d			1.3	1.1		
Total		10.1	37.1	1.3	4.7	11.3	41.8
3	3a	9.7	18.5	0.8	1.5	10.5	20.0
	3b	27.9	50.7	1.4	2.6	29.3	53.4
Total		18.5	69.3	1.1	4.1	19.6	73.4
4	4a			0.8	1.0		
	4b			0.3	0.4		
	4c			0.6	1.1		
	4d			0.8	1.2		
Total		0.6	3.6	0.6	3.7	1.2	7.3
5	5a			0.5	0.8		
	5b			0.9	3.1		
Total		20.6	101	0.8	3.9	21.3	105
All		10.9	247	1.2	28.4	12.0	275

and 247 Gmol N yr^{-1} as nitrate and ammonium respectively to the Atlantic (Tables 5 and 6).

3.2. Dry Deposition: Chemical Characteristics of Air Mass Types

[16] In Figure 4 and Table 7 we summarize the characteristic nitrogen species concentrations of each of the 8 air mass types we identified. Aerosol soluble inorganic nitrogen in our data set is composed of ~53% nitrate and ~47% ammonium (median values for all data), with ~90% and ~14% respectively of each species being present on coarse mode aerosol. These proportions and size distributions, and the concentrations observed in individual air mass types, are very similar to those we reported earlier for a subset of the data described in this manuscript [*Baker et al.*, 2006a]. For most air mass types, median concentrations of nitrate and ammonium varied by less than 20% between the two data sets, while the number of observations increased by 1.5 – sixfold.

[17] Our data are also consistent with other results for specific air mass types (see Table 8). The relatively high concentrations in the Northern Hemisphere air mass types emphasize the importance of industrial and agricultural emissions to the atmospheric nitrogen budget, but our results also illustrate significant inputs to the South Atlantic, associated with biomass burning [*Swap et al.*, 1996; *Baker et al.*, 2006a].

[18] Although our sample set is relatively large, the inherent variability of the atmosphere means that simple averages of results in any particular area may not be wholly representative of the long-term average, either because sampling may have taken place during periods of relatively atypical airflow, or because deposition from atypical airflows constitutes a significant proportion of total deposition [e.g., *Spokes et al.*, 2000]. To overcome this potential bias, we have weighted average aerosol concentrations in each ocean region to reflect the climatological average airflow, and the methods used to do this are now described.

3.3. Dry Deposition: Air Mass Climatology

[19] The airflow in each region was characterized using daily 5-day air mass back trajectories obtained over a 5 year period for points located roughly in the center of each box (see Figure 3). Each daily back trajectory was then assigned to one of the air mass types identified above, based on which source regions the trajectories had contact with, and this information was compiled into a flow climatology for the Atlantic (see Appendix A). The resulting climatology is consistent with the well-established broad flow patterns of the Atlantic, but goes further by identifying the frequency of flow from both the dominant and minor source regions.

[20] Although we identified an air mass back trajectory type that appeared to be associated with austral spring
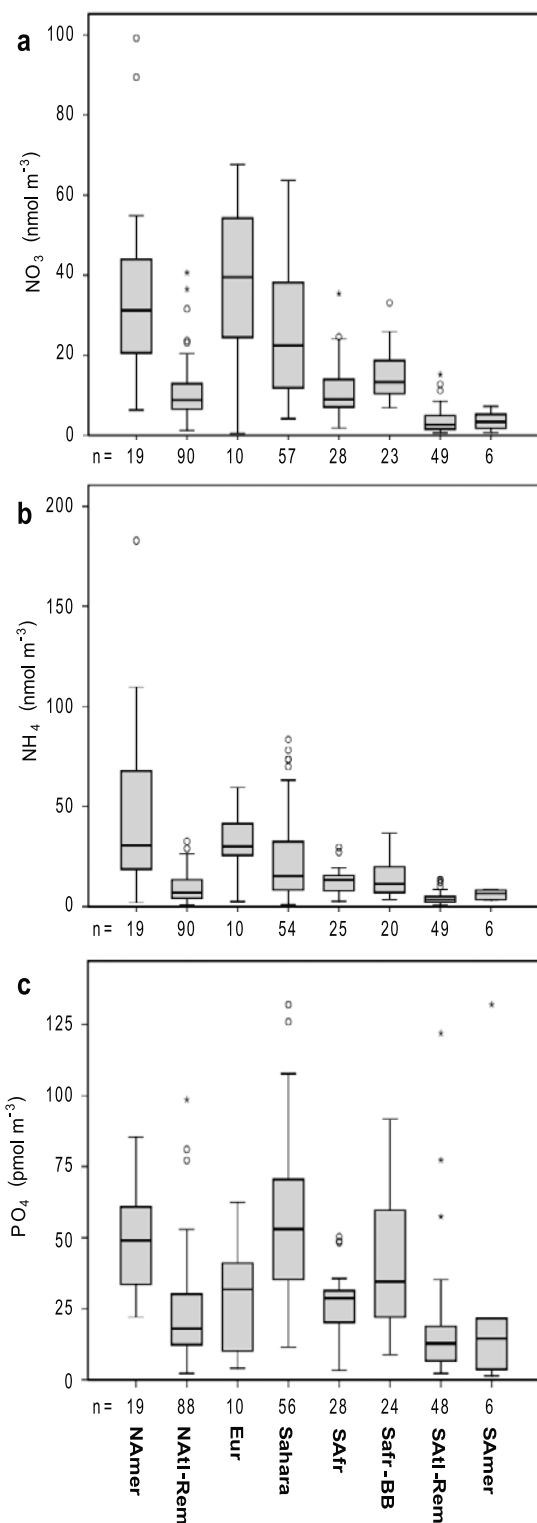
Figure 4. Box and whisker plots, showing total (coarse + fine) concentrations of (a) nitrate, (b) ammonium and (c) soluble phosphate according to air mass type. Panels show the number of observations (n) in each category. Air mass types are defined in the text.

biomass burning in southern Africa in our cruise data, we found that this back trajectory type occurred throughout the year in our air mass climatology. In order to include this biomass burning in our climatology, whenever that back trajectory type was identified during the biomass burning season in the region (April–October [*Cooke et al.*, 1996]) we assigned that day to the SAfr-BB type in the climatology. During the rest of the year such air mass back trajectories were assigned to the SAfr type. For all the back trajectory points in the Southern Hemisphere the annual climatology was therefore a time-weighted average of the climatologies for the biomass burning and non-biomass burning seasons (Table A1).

[21] The flux into each dry deposition sub-region ($F^{d,int}$) was calculated as the sum of the flux associated with flow from each source region (F_i^d), weighted by the frequency of transport (w_i^{am}; see Table A1) from that sector (equation 4). Aerosol concentrations used in this calculation were the median for all samples of each air mass type collected within each large (wet) deposition region. In this way we sought to maximize the available data for each air mass type, but retain a degree of spatial resolution so that concentrations varied with proximity to source. Thus, for example, we used NO_3 concentrations of 7.8, 35.4, 11.8 and 24.8 nmol m^{-3} when computing the dry flux contribution from Saharan air in Regions 1, 2, 3a and 3b (see Table 7). We did not attempt to account for variations in concentration within each region.

$$F^{d,\text{int}} = \sum w_i^{am} F_i^d \qquad (4)$$

[22] As for the wet deposition estimate, where samples were below detection limit, concentration values equal to

Table 7. Median Concentrations and Number of Observations (in Parentheses) for Nitrate, Ammonium, and Phosphate for the Aerosol Samples Collected During This Work Divided According to Air Mass Type in Each of the Five Broad Deposition Regions

Region	Air Mass	NO_3 (nmol m^{-3})	NH_4 (nmol m^{-3})	PO_4 (pmol m^{-3})
1	NAmer	31.2 (19)	30.5 (19)	48.7 (19)
	NAtl-Rem	7.3 (31)	6.5 (31)	19.7 (31)
	Sahara	7.8 (2)	4.4 (2)	17.1 (2)
2	NAtl-Rem	9.3 (58)	7.5 (58)	19.0 (56)
	Eur	39.5 (10)	30.1 (10)	29.1 (10)
	Sahara	35.4 (26)	18.8 (24)	61.5 (26)
3a	NAtl-Rem	9.2 (1)	22.4 (1)	22.6 (1)
	Sahara	11.8 (17)	9.8 (17)	34.8 (17)
	SAfr	6.3 (4)	5.6 (4)	17.4 (4)
	SAfr-BB	8.8 (3)	11.0 (3)	25.4 (3)
3b	Sahara	24.8 (12)	15.1 (11)	52.6 (11)
	SAfr	9.3 (20)	13.7 (18)	29.6 (20)
	SAfr-BB	11.0 (7)	8.2 (6)	41.0 (7)
	SAtl-Rem	7.4 (1)	11.9 (1)	21.9 (1)
4	SAfr	9.5 (4)	13.2 (3)	17.9 (4)
	SAfr-BB	14.8 (14)	12.4 (12)	34.2 (14)
	SAtl-Rem	2.7 (33)	3.3 (33)	12.8 (33)
5	SAtl-Rem	2.8 (14)	3.7 (14)	12.4 (14)
	SAmer	3.4 (6)	6.4 (6)	14.6 (6)

Table 8. Examples of Previously Reported Concentrations and Number of Observations (in Parentheses, Where Available) for Nitrate, Ammonium, and Phosphate for Aerosol Samples Divided According to Air Mass Type in Each of the Five Broad Deposition Regions Used in This Work

Region	Air Mass	NO_3 (nmol m^{-3})	NH_4 (nmol m^{-3})	PO_4 (pmol m^{-3})	Reference[a]
1	NAmer	5.1–52 (5)	9.3–64 (5)		A
	NAmer	38[b]	30[b]		B
	NAmer			23–200 (7)	C
	NAtl-Rem	3.9–6.1 (2)	5.2–7.0 (2)		A
	NAtl-Rem	3.9[b]	3.9[b]		B
	NAtl-Rem			6.5–19 (2)	C
	?			6.5–310 (24)	C
2	NAtl-Rem	4.2[b]	9.7[b]		D
	Eur	82[b]	112[b]		D
	Eur	22[b]	24[b]		E
	Eur	20.8–37.3			F
	Sahara	14.2–29.9			F
	Sahara	2.1–21.6 (5)	6.5–44.2 (5)		A
1/2/3a	?			10–2890 (89)	G
3a	Sahara	13[b]	5[b]		B
3b	SAfr-BB	15[b]	11[b]		B
4	SAfr	3.1[b]	2.8[b]		B
	SAfr	5.7–16.8			F
	SAfr & SAtl-Rem	3.7[b]	3.9[b]		E
	SAtl-Rem	2.3[b]	2.2[b]		B
	SAtl-Rem	0.3[b]	1.8[b]		E

[a]References: A, *Church et al.* [1991]; B, *Quinn et al.* [2001]; C, *Graham and Duce* [1982]; D, *Spokes et al.* [2000]; E, *Virkkula et al.* [2006]; F, *Keene et al.* [2009]; G, *Chen* [2004].

[b]Average value.

75% of detection limit were used. This most often applied to ammonium in coarse mode aerosol or nitrate in fine mode aerosol (i.e., the modes in which they are least abundant) and hence the overall nitrogen concentration was well above detection limit. Since we used median values for the calculation of deposition fluxes for each air mass type, the inclusion of these below detection limit values had a negligible effect on our dry flux estimates.

[23] Our estimate of dry nitrate and ammonium input to the Atlantic is 136 Gmol N yr^{-1} and 28 Gmol N yr^{-1} (Tables 5 and 6) respectively.

3.4. Contribution of Soluble Organic Nitrogen

[24] There is increasing evidence that soluble organic nitrogen (SON) species make up a significant fraction of atmospheric reactive nitrogen [*Cornell et al.*, 1995; *Duce et al.*, 2008; *Zhang et al.*, 2008; *Lesworth et al.*, 2010], and we have strong evidence for the presence of SON in some of the samples used in this study [e.g., *Baker et al.*, 2007; *Lesworth et al.*, 2010]. Because of the diverse nature of SON constituents, overall SON concentrations are generally determined by subtracting the sum of nitrate and ammonium from total soluble nitrogen in each sample, and this is the method that we have used for our data. However, as discussed in detail by *Cornell et al.* [2003], the accurate determination of SON concentrations is hampered by problems associated with the large error resulting from the three individual N determinations necessary to calculate SON, and potentially

also by changes in ammonium concentrations in samples if the individual N species are not analyzed concurrently. For some of the cruises reported here, a significant fraction of our calculated SON data had a magnitude lower than their respective standard deviation, or were negative, which indicated problems with the analysis. We therefore decided to treat the estimation of SON fluxes differently from those of the other N species. We use our current and previous results for the fraction of soluble N present as SON, and those reported in other studies, to estimate that SON represents 13–29% of N in marine aerosol, with 40% of this SON in coarse mode aerosol [*Spokes et al.*, 2000; *Nakamura et al.*, 2006; *Baker et al.*, 2007; *Lesworth et al.*, 2010], and 14–52% of N in rainfall [*Spokes et al.*, 2000; *Neff et al.*, 2002]. Using these values we calculate that SON contributes 19–52 Gmol N yr^{-1} in dry deposition and 94–625 Gmol N yr^{-1} in wet deposition to the Atlantic.

3.5. Dry Deposition of Phosphate

[25] For all of the cruises discussed above we determined soluble phosphate concentrations, as well as soluble nitrogen concentrations, in the aerosol samples collected [e.g., *Baker et al.*, 2006a, 2007]. We did not measure phosphate concentrations in the rainfall samples collected. Table 7 and Figure 4 illustrate the characteristic phosphate concentrations for each air mass type. There are very few reports of aerosol soluble phosphorus concentrations over the Atlantic Ocean. However, our results compare well with most other

Table 9. Dry Deposition Fluxes and Dry Annual Inputs of Phosphate to the Five Broad Deposition Regions and the Molar Ratio of Total (Inorganic Plus Soluble Organic) N:P in Dry Deposition

Region	Sub-region	Dry Flux (μmol m^{-2} d^{-1})	Dry Input (Gmol yr^{-1})	N:P
1	1a	0.017	0.006	1260–1490
	1b	0.016	0.012	1120–1310
	1c	0.015	0.017	1260–1480
	1d	0.011	0.011	990–1150
	1e	0.009	0.011	870–1000
Total		0.013	0.058	1100–1290
2	2a	0.010	0.007	1360–1590
	2b	0.009	0.008	1420–1650
	2c	0.009	0.012	990–1160
	2d	0.015	0.013	1320–1520
Total		0.011	0.040	1240–1450
3	3a	0.007	0.013	990–1200
	3b	0.009	0.016	770–910
Total		0.008	0.029	870–1040
4	4a	0.007	0.009	1170–1420
	4b	0.003	0.004	1090–1320
	4c	0.005	0.008	770–920
	4d	0.006	0.008	670–790
Total		0.005	0.030	910–1090
5	5a	0.005	0.007	610–730
	5b	0.006	0.021	630–760
Total		0.006	0.028	620–750
All		0.008	0.185	990–1170

previous data, although they are significantly lower than those of *Chen* [2004] (as cited by *Mahowald et al.* [2008]) in the tropical North Atlantic (Table 8).

[26] We use the aerosol phosphate data to estimate the climatological dry deposition of phosphate to the Atlantic in the same manner described above for inorganic nitrogen inputs. Dry phosphate flux to each region is shown in Table 9. In total we estimate the Atlantic receives ~0.19 Gmol yr^{-1} of phosphate in dry deposition, 69% of which enters our study region north of 5°S.

4. Discussion

[27] Our results for nitrogen are summarized in Tables 5, 6, and 10. We estimate total nitrogen deposition in the Atlantic from 50°N to 50°S to be 854–1420 Gmol N yr^{-1}, with ~78–85% of this in the form of wet deposition, although this is highly variable between regions (Table 10). Previous estimates of the proportion of atmospheric nitrogen input in wet deposition, summarized by *Jickells* [2006], are in the range 55–79%. The majority of this deposition occurs to the north of 5°S, reflecting presumably the greater anthropogenic inputs of nitrogen to the Northern Hemisphere. Our results suggest that nitrate is probably the most abundant nitrogen species in deposition (Table 10), largely due to the more efficient deposition of coarser nitrate-containing aerosols. Our estimate of dry soluble phosphorus deposition to the same area also indicates that inputs are greater to the Northern Hemisphere, with deposition fluxes being highest to the regions adjacent to North Africa and North America (Table 9). Below we discuss potential sources of bias and uncertainty in these estimates and compare our results to other estimates of atmospheric nitrogen and phosphorus inputs to the Atlantic.

4.1. Seasonal Effects on Climatology

[28] Although our air mass weighting approach avoids biases associated with changes in airflow, our sampling in this work has been largely based on inter-hemispheric transfers of polar research vessels and such transfers are necessarily restricted to the spring/autumn seasons. Our climatology is therefore subject to potential biases associated with seasonally variable atmospheric sources or deposition processes (although we do not consider the latter here). *Prospero et al.* [1996] discussed seasonality in the major nitrogen sources to the atmosphere around the North Atlantic. They noted that while there was some seasonality in oxidized nitrogen emissions from power generation (highest in winter)

Table 10. Summary of Annual (Wet Plus Dry) Nitrogen Inputs From Nitrate, Ammonium, and SON to the Atlantic and the Percentage of Total N Input to Each Region in Wet Deposition[a]

Region	Nitrate (Gmol yr^{-1})	Ammonium (Gmol yr^{-1})	SON (Gmol yr^{-1})	Total N (Gmol yr^{-1})	% Total N in Wet Deposition
1	157	47.3	30.2–176	235–381	73–80
2	77.2	41.8	16.7–93.1	136–212	64–73
3a	42.6	20.0	9.9–59.4	72.5–122	82–87
3b	43.0	53.4	15.2–96.3	112–193	89–93
3	85.6	73.4	25.1–156	184–315	86–90
4	31.3	7.3	5.6–24.7	44.2–63.3	39–49
5	115	105	35.4–228	256–448	93–95
All	466	275	113–677	854–1420	78–85

[a]Nitrate and ammonium data are taken from Tables 5 and 6, respectively. SON data are estimated as a fixed fraction of total inorganic N inputs, as described in section 3.4.

and reduced nitrogen from agricultural activity (highest in spring/summer), the variations in emission were rather small relative to the overall nitrogen emission. *Hastings et al.* [2003] noted no significant difference in the VWM nitrate concentrations in rainfall collected during cool (October–March) and warm (April–September) seasons at Bermuda. Similar considerations probably apply to seasonal variations in emissions from power generation and agricultural sources in the Southern Hemisphere. In the tropics there are also seasonal sources of atmospheric nitrogen associated with biomass burning. In the region affected by southern African biomass burning we have samples from both burning and non-burning seasons, but we have very few samples representative of the biomass burning season in the Sahel, which occurs through the months of November to February [*Cooke et al.*, 1996]. *Quinn et al.* [2001] reported average nitrate and ammonium concentrations of ~15 nmol m^{-3} and ~10 nmol m^{-3} respectively in biomass burning-impacted aerosols over the equatorial Atlantic in January/February 1999. These values are generally higher than we observed in non-dusty, non-biomass burning-impacted aerosols in that region (see Table 7, SAfr type, Regions 3a and 3b), although the difference is less than a factor of 3. Overall, we argue that seasonal changes in nitrogen emissions are probably not an important source of uncertainty, except perhaps for areas affected by biomass burning. Seasonality in sources of soluble phosphorus is probably dominated by seasonality in mineral dust generation, which is highest in the southern Sahara and Sahel in boreal spring/summer and in the, much weaker, Southern Hemisphere dust sources in austral spring/summer [*Werner et al.*, 2002]. Thus seasonal variations are likely to introduce greater uncertainty into our estimate of soluble phosphorus inputs than was the case for nitrogen inputs. As for nitrogen emissions, we probably capture the signal of phosphorus emissions from biomass burning in southern Africa, but miss the signal from biomass burning in the Sahel. Because of the possibility that we did not sample periods of more intense mineral dust transport, particularly from the Sahara/Sahel, we consider that our soluble phosphorus input estimates should be considered as lower limits.

4.2. Other Sources of Uncertainty in Our Flux Estimates

[29] Although we have a relatively large database of aerosol observations on which to base our dry input estimates, once sub-divided into deposition regions and air mass types the number of observations can become rather small (Table 7). Thus, in some cases, our median concentrations may not be representative of the aerosol populations we seek to characterize. We explore the potential effect of this by comparing our median air mass type concentrations with those we reported previously for the first four of the cruises used here [*Baker et al.*, 2006a]. The differences between the median concentrations for each air mass type between the two data sets decrease with increasing number of observations in the earlier data set. We therefore attempt to account for the uncertainty in characterizing median air mass concentrations by assigning different uncertainties to each concentration depending on

the number of observations used to calculate the median, with the values chosen for these uncertainties (i.e., ±100% for n ≤ 2, ±50% for 3 < n ≤ 5, ±30% for 6 < n ≤ 10 and ±15% for n > 10) being based on the results of the comparison mentioned above. Propagation of these uncertainties through our air mass weighted flux calculation (equation 4) results in uncertainties of 15, 18, 53, 32 and 21% for dry inorganic nitrogen input to Regions 1–5 respectively and corresponding values of 15, 16, 51, 28 and 20% for dry phosphate input. Overall we estimate the uncertainties in our dry input values to be 26% for both inorganic nitrogen and phosphate. This analysis, however, does not take account of biases introduced by the non-uniform distribution of samples through our deposition regions.

[30] We also compare the proportions of observations of each air mass type in each region with the frequency of occurrence of that air mass type in our air mass climatology, in an attempt to assess whether we have under- or over-sampled air from any particular source. This analysis indicates that in several regions (1, 2 and 5) there is very good agreement between the distribution of samples between air mass types and the occurrence of these air mass types in the climatology. In Region 3 the agreement is poor, particularly in the west (R3a) where we have significantly more samples designated as Saharan than were identified as such from back trajectory analysis in the climatology. This effect is probably caused by our use of aerosol color to classify samples as Saharan (see Section 2.2). Transport times for Saharan dust may be longer than 5 days to the west Atlantic and so our air mass climatology (based on 5 day back trajectories) probably underestimates the occurrence of Saharan air in the west Atlantic. In Region 3b the climatology predicts a much greater proportion of SAtl-Rem air masses than observed in our database. This may be due to zonal differences in migration of the ITCZ across this region. We estimate that these systematic biases lead to underestimations of total inorganic nitrogen input of 3% and 14% to Regions 3a and 3b respectively, and of 8% to Region 3 as a whole and 1% to the Atlantic overall. Corresponding values for the underestimation of phosphate inputs to these regions due to these systematic biases were 18, 9, 13, and 2% respectively. In Region 4 the climatology predicts proportionally more SAfr type air and less SAfr-BB type air than observed in our sample set. This is probably a result of the seasonal bias in our sampling program, since 6 of the 9 cruises to pass through the South Atlantic occurred during the austral spring biomass burning season.

[31] Dry deposition velocities remain one of the major uncertainties in assessing the atmospheric flux of material to the ocean [*Arimoto et al.*, 2003]. The uncertainty in the aerosol dry deposition velocity is especially large for particles that have a mass-size distribution in the transition from the accumulation- to coarse mode range (~0.5 − 5 μm). This is due to a strong increase in v$_d$, especially for relative large wind speeds, for this aerosol radius range [*Ganzeveld et al.*, 1998]. Ideally measured mass-size distributions should be used to constrain values of v$_d$, but we are not able to do this for our data set. Other main uncertainties in v$_d$ include the considered growth effect associated with high humidity and the enhanced removal due to the presence of whitecaps.

We base our estimate of the uncertainty associated with variation in v_d due to wind speed and particle size for the coarse size range. For our chosen coarse particle size (7 μm), use of the minimum and maximum climatological wind speeds for the Atlantic yields values of v_d ~45% lower and ~88% higher respectively than those used in our baseline calculation. We assess the influence of particle size by calculating v_d for particles of 5 and 9 μm at a wind speed of 7 m s^{-1} (our mean climatological wind speed). This yields values of v_d ~12% lower and ~31% higher respectively than those used in our baseline calculation. Overall we estimate the range of values for total inorganic nitrogen inputs in dry deposition to the Atlantic due to these uncertainties in deposition velocity to be $72 - 318$ Gmol N yr^{-1} respectively, compared to 165 Gmol N yr^{-1} for our baseline calculation. The uncertainty in deposition velocities is therefore the dominant source of uncertainty in our dry input estimates.

[32] We have far fewer rainfall samples than aerosol samples and we have attempted to compensate for this by considering wet deposition on a coarser scale than we did for dry deposition. In general, we consider that the number and distribution of rainfall samples available to us (Table 4) is probably too limited to be truly representative of rainfall over the Atlantic. In Section 3.1 we compared our rainwater nitrogen concentration data to other published values. That comparison emphasized the sparcity of data available for the Atlantic, particularly, and inevitably, for the drier zones (Regions 2 and 4). For a few published studies VWM concentrations for nitrate and ammonium, or enough information to calculate this parameter, are given and we use this information to try to assess the likely uncertainty in our wet input estimates. For nitrate in Region 1, data are available at Bermuda from two long-term studies (1980–88, C_R = 4.60 μmol L^{-1} and 1987–97, C_R = 4.57 μmol L^{-1}; J. N. Galloway, personal communication, 2009) and a more recent yearlong study (2000–01, C_R = 5.0 μmol L^{-1} (April–September) and C_R = 6.4 μmol L^{-1} (October–March); [Hastings et al., 2003]). A long-term record is also available for Barbados, but we do not use this data here as the dominant flow to Barbados is of maritime air, rather than the North American outflow which affects most of Region 1. Ammonium data are also available for the long-term studies at Bermuda (1980–88, C_R = 3.43 μmol L^{-1} and 1987–97, C_R = 2.55 μmol L^{-1}; J. N. Galloway, personal communication, 2009). We use the values most different to our own VWM concentrations for the region (4.57 μmol L^{-1} for nitrate and 3.43 μmol L^{-1} for ammonium) to indicate the maximum potential difference in wet nitrate input to Region 1. This results in inputs 44% lower and 33% higher than the values given in Tables 5 and 6 for nitrate and ammonium respectively and 25% lower for total inorganic nitrogen. We are not aware of any long-term studies in the other regions, but Losno et al. [1991] reported sample volumes and nitrate concentrations for 4 rain events collected in our Region 3a and 5 events in our Region 5. We combine their results with ours to recalculate VWM nitrate concentrations in those regions and derive wet nitrate inputs 9% lower in Region 3a and 20% higher in Region 5 than the values given in Table 5. Overall we consider the uncertainty in our wet (inorganic) nitrogen input estimates

to be ~20% for the high precipitation regions (1, 3 and 5). In the low precipitation regions (2 and 4) the uncertainty is higher (we estimate ~40%) because rainfall in these regions is chronically under-sampled and because the few samples we do have in these regions come from their margins, where precipitation rates are higher, so may not be representative of the region as a whole. Because wet deposition in the dry regions contributes relatively little to the total, we estimate the overall uncertainty in our wet input value to the study region to be ~25%.

[33] There is also a potential bias introduced into our wet input calculation due to the precipitation field that we have used [Xie and Arkin, 1997]. However, while other precipitation fields [e.g., Elliott and Reed, 1984; Yuan and Miller, 2002] differ slightly in the distribution of precipitation they agree relatively well in terms of total rainfall amount. We therefore consider that the uncertainty arising from choice of precipitation field is relatively minor compared to that in species concentration data discussed above, particularly over the broad spatial scales used here.

4.3. Comparison to Other Flux Estimates

[34] In Table 11 we compare our estimates of atmospheric inputs to the Atlantic to some other recent studies [Duce et al., 1991; Prospero et al., 1996; Dentener et al., 2006; Luo et al., 2007; Mahowald et al., 2008]. Where possible we have attempted to compare inputs to similar areas to those used in our study.

[35] Duce et al. [1991] made their estimates on the basis of extrapolation of land-based observations and measurements over the oceans using constant dry deposition velocity values of 0.1 and 0.3 cm s^{-1} for ammonium and nitrate respectively, and quoted results for the whole North and South Atlantic. Although they did not specify the boundaries of these regions, the total areas covered were given as 54.9×10^{12} m^2 and 51.2×10^{12} m^2 for the North and South Atlantic respectively, compared to the 33.0×10^{12} m^2 (Regions 1–3) and 29.7×10^{12} m^2 (Regions 4 and 5) used here. Hence their results actually correspond to smaller inputs to the regions covered by our study, although the magnitude of the reduction in input is probably not a simple function of the relative surface areas used because of the spatial variability in atmospheric fluxes. Prospero et al. [1996] used the average of 5 diverse models to calculate atmospheric inputs to 10° x 10° grid cells of the non-coastal North Atlantic. We have summed the values given for the cells in the region 0–50°N, 80°W–10°E to calculate the values given in Table 11. The other studies all use results from emissions/atmospheric chemical transport models [Dentener et al., 2006; Luo et al., 2007; Mahowald et al., 2008] and the lead authors have kindly provided us with values for the basin between 50°S and 50°N (excluding the Caribbean), with the north/south division at ~5°S. None of the studies which considered nitrogen deposition estimated organic nitrogen inputs explicitly, so we restrict our comparison to inorganic nitrogen species.

[36] For total inorganic nitrogen input, there is generally good agreement between the values listed in Table 11, although our results for nitrate appear to be slightly higher, and for ammonium slightly lower, than those for similar

Table 11. Comparison of Inorganic Nitrogen and Soluble Phosphorus Input Estimates to the North, South, and All Atlantic Ocean and the Ratio of Inputs to the North and South Basins (N/S)[a]

Reference	Species	N. Atl. (Gmol yr^{-1})	S. Atl. (Gmol yr^{-1})	N/S	All Atl. (Gmol yr^{-1})
Duce et al. [1991][b]					
	NO$_y$	326	46	7.1	373
	NH$_x$	292	68	4.3	360
	TIN	619	114	5.4	733
	Sol P[b,c]	0.59	0.06	9.8	0.65
Prospero et al. [1996][d]					
	NO$_y$	237			
	NH$_x$	173			
	TIN	409			
	Sol P[b,c]	0.45			
Dentener et al. [2006][e,f]					
	NO$_y$	312	113	2.8	425
	NH$_x$	180	131	1.4	311
	TIN	492	244	2.0	736
Luo et al. [2007][e,g]					
	NO$_y$	265	120	2.2	384
	NH$_x$	197	156	1.3	352
	TIN	462	275	1.7	737
Mahowald et al. [2008][e,h]					
	Sol P[i]	0.22	0.08	2.7	0.31
This work[j]					
	NO$_3$	320	146	2.2	466
	NH$_4$	163	113	1.4	275
	TIN	483	259	1.9	741
	Sol P[j]	0.13	0.06	2.2	0.19

[a]Nitrogen estimates are divided into contributions from nitrate or oxidised nitrogen (NO$_y$), ammonium or reduced nitrogen (NH$_x$) and total inorganic nitrogen (TIN).
[a]Total area North Atlantic (incl. Caribbean) 54.9×10^{12} m^2, South Atlantic 51.2×10^{12} m^2.
[b]Only input of P in mineral dust was considered in this study.
[c]Wet plus dry deposition, calculated assuming 8% of P in dust is soluble.
[d]Area is the Atlantic between 0°N–50°N, 80°W–10°E.
[e]Area is the Atlantic between 50°S and 50°N, excluding the Caribbean, divided at 5°S.
[f]F. J. Dentener, personal communication, 2009.
[g]C. Luo, personal communication, 2009.
[h]N. M. Mahowald, personal communication, 2009.
[i]Dry deposition of soluble P only.
[j]North Atlantic is sum of Regions 1–3, South Atlantic is sum of Regions 4 and 5.

areas of the Atlantic. All the previous studies that gave estimates in both hemispheres predicted a much stronger north–south gradient for NO$_y$ than for NH$_x$, and our results are in agreement with this (Table 11). Apart from the study of *Duce et al.* [1991], whose estimate for the South Atlantic was significantly lower than those of the other studies, our ratios of nitrogen input to the North and South Atlantic are similar to those of the other studies, suggesting good agreement between the broad-scale distribution of nitrogen deposition.

[37] When considering atmospheric phosphorus inputs, *Duce et al.* [1991] and *Prospero et al.* [1996] calculated total phosphorus deposition from estimates of mineral dust deposition, assuming that mineral dust had the same phosphorus content as average crustal material. The soluble phosphorus input values given in Table 11 for these two studies have been calculated from these total phosphorus inputs assuming that 8% of phosphorus in mineral dust is soluble [*Baker et al.*, 2006b]. Mineral dust is the dominant source of total and soluble phosphorus to the atmosphere [*Mahowald et al.*, 2008], but for soluble phosphorus other sources, such as biomass burning and anthropogenic emissions, are relatively more important [*Baker et al.*, 2006a; *Mahowald et al.*, 2008]. Our calculations of soluble phosphorus inputs from the *Duce et al.* [1991] and *Prospero et al.* [1996] studies (Table 11) are therefore probably slight underestimates because they do not consider these other sources. *Duce et al.* [1991] and *Prospero et al.* [1996] also give their estimates only as total (wet plus dry) deposition. We convert their values to dry input by assuming that the relative contributions of wet and dry deposition are the same for phosphorus as our results for nitrogen (i.e., ~15–25% dry deposition (Table 10)). The calculated dry soluble phosphorus inputs are in the range 0.09–0.15 Gmol P yr^{-1} and 0.07–0.11 Gmol P yr^{-1} for the *Duce et al.* [1991] and *Prospero et al.* [1996] estimates in the North Atlantic respec-

tively, in comparison to the values of 0.22 Gmol P yr^{-1} from *Mahowald et al.* [2008] and our result of 0.13 Gmol P yr^{-1}. Considering the caveats regarding the *Duce et al.* [1991] and *Prospero et al.* [1996] estimates (above) and the seasonal bias in our data set (Section 4.1), there is reasonably good agreement between these values. In terms of the north-south distribution of phosphate inputs, *Mahowald et al.* [2008] predict a stronger inter-hemispheric gradient than indicated by our results, and this might, at least in part, be consistent with our under-sampling of mineral dust exported from North Africa. Again the *Duce et al.* [1991] estimate for the South Atlantic is very much lower than the other results.

4.4. N:P Ratios in Dry Deposition

[38] The molar ratio of total (inorganic and soluble organic) N:PO$_4$ inputs in dry deposition in our climatology overall is in the range 990–1170 (Table 9). This is consistent with previous estimates of this parameter in aerosol collected over the Atlantic [*Baker et al.*, 2003; *Duarte et al.*, 2006; *Baker et al.*, 2007] and, since rainfall components (and P in particular) are largely derived from aerosol, the N:P ratio of total deposition is likely similar to, or higher than, this range. This ratio indicates that atmospheric nitrogen supply is significantly enriched relative to phytoplankton requirements (N:P ~8–45 [*Arrigo*, 2005]) over the whole region. It is also apparent from Table 9 that there is a strong inter-hemispheric gradient in atmospheric N:P ratio, with significantly higher values in the Northern Hemisphere than in the Southern Hemisphere. This is consistent with our understanding of the relative rates of nitrogen emissions in the two hemispheres and also with there being a greater anthropogenic enhancement in atmospheric phosphate concentrations over the South Atlantic [*Mahowald et al.*, 2008]. In agreement with previous studies [*Baker et al.*, 2003, 2007], the implication of the high N:P ratios in our dry deposition estimates is that the atmosphere cannot be a source of "new" phosphorus to the oceans, as the deposition of phosphorus is always accompanied by a stoichiometric excess of atmospheric nitrogen.

5. Conclusion

[39] We use results of rainfall and aerosols sampling during a dozen basin-scale transect cruises to estimate atmospheric inputs of nitrogen species and soluble phosphorus to the Atlantic Ocean. These estimates are subject, by varying degrees, to uncertainties associated with poor spatial distribution of data (particularly for wet deposition in the drier regions of the basin), inadequate characterization of the representative concentrations used in the calculations, seasonal bias in our sampling program and poorly constrained dry deposition velocities. We consider our dry phosphate input estimate to be a lower limit, as our sampling program likely missed periods of enhanced atmospheric phosphorus transport associated with outflow of Saharan dust. Nevertheless our results agree well with previous estimates of inorganic nitrogen and soluble phosphorus atmospheric inputs to the Atlantic (Table 11) and provide a significant new observational basis with which to constrain modeling

studies of atmospheric nutrient input to the region. Given the uncertainties in dry deposition fluxes, we consider that comparisons between field observations and model output in studies of this type would be better done using concentration, rather than deposition, fields. Atmospheric nitrogen inputs to the Atlantic appear to be greater in wet deposition than dry, which suggests that further rain sampling, particularly in the Southern Hemisphere and drier regions of the basin, would be the most useful sampling activity to better constrain our estimate.

[40] In addition to our results for inorganic nitrogen, we made a more tentative estimate of soluble organic nitrogen input to the Atlantic, which increases the total nitrogen input by 13–47%. This suggests that SON may make a significant contribution to atmospheric nitrogen inputs to the Atlantic, but our understanding of the occurrence, distribution, chemistry and fate of atmospheric SON is still poor.

Appendix A: Dry Deposition Air Mass Climatology

[41] Air mass types were assigned for each day of the 5 year period 2001–2005, based on the positions of air parcels at the 3 arrival heights (50 m, 500 m and 1000 m above sea level) at both 3 and 5 days upwind of each of the air mass climatology points 1a–5b (see Figure 3 and Table A1). These air mass types correspond to the major aerosol sources regions around the Atlantic, as described in Section 2.2. Three altitudes were used because aerosols can be transported from a source region at high altitude followed by subsequent gravitational settling or mixing of these aerosols into lower altitude air masses which may not have been near the source region. This is particularly important for high altitude transport of Saharan dust and aerosols from southern Africa [*Carlson and Prospero*, 1972; *Tyson et al.*, 1996].

[42] Generally the aerosol source region/air mass type for each day was assigned on the following basis: In most cases, the surface (50 m) trajectory probably represented the dominant air parcel, especially if the 500 m and 1000 m trajectories were from a remote region. In such cases, the surface trajectory, for both 3 and 5 days, was used to identify the source region. However, if both the 3 day 500 m and 1000 m trajectories were from a continental source region and the 50 m trajectory was from a remote region, the continental region was taken as the source region. For the 5 day classification the surface trajectory was taken as the source region in all cases, except when one of the higher level trajectories was from an African source region, where we consider high-level transport to be the dominant process, as discussed above. Finally, the air mass type was assigned as that indicated by the source region identified from the 3 day back trajectory position, unless this was from a remote region and the 5 day source region was from a more polluted region, in which case the 5 day source region was taken to represent the air mass type.

[43] This approach worked well for all regions, except in the tropical and eastern South Atlantic (points 3a2, 3b, 4a, 4b, 4c and 4d). Here we observed samples with concentrations significantly above background for continentally derived tracers [*Baker et al.*, 2006a], but for which 5 day back trajectories often approached, but rarely touched,

Table A1. Percentage Occurrence of Different Air Mass Types (w_i^{am}) at Selected Points in the Atlantic, Based on Analysis of Daily 5-Day Air Mass Back Trajectories for the Period 2001–2005[a]

	Location	NAmer	NAtl- Rem	Eur	Sahara	SAfr	SAfr-BB	SAtl- Rem	SAmer
1a	44°N 56°W	55.5	44.5	0.1					
1b	44°N 38°W	32.7	63.6	3.4	0.3				
1c	32.4°N 64.7°W	54.7	45.3						
1d	32.4°N 38°W	16.9	79.3	2.1	1.7				
1e	20°N 56°W	6.0	92.4		1.6				
2a	44°N 16°W	13.0	70.0	16.3	0.7				
2b	32.4°N 16°W	4.7	68.5	16.8	10.0				
2c	20°N 44°W	3.4	86.9	0.5	9.1			0.2	
2d	20°N 20°W	0.7	42.6	5.9	50.7			0.1	
3a1	10°N 32°W		47.9	0.4	46.2			5.6	
3a2	0°N 32°W		3.9/11.9		1.0/8.6	67.0/75.2	23.0/-	5.1/4.3	
3b	2.25°N 2.1°W		0.2/0.4		3.8/13.0	43.8/28.6	8.6/-	43.6/58.1	
4a	10°S 25°W					83.1/100	14.2/-	2.7/-	
4b	10°S 4°W					40.3/85.7	32.3/-	27.4/14.3	
4c	25°S 4°W					14.1/12.7	5.3/-	79.9/87.3	0.7/-
4d	34°S 12°E					3.2/2.1	1.6/-	93.6/97.2	1.6/0.7
5a	25°S 36°W						9.6/-	75.4/95.0	14.9/5.0
5b	37.5°S 41.7°W						3.0/-	50.1/62.6	46.9/37.4

[a]For points in the Southern Hemisphere, values are given for the biomass-burning and non-biomass-burning seasons, respectively.

southern Africa. Thus the method described above was not always able to distinguish the SAfr and SAfr-BB air mass types from the SAtl-Rem type. We used the following means to identify these air mass types, based on the characteristics of the air mass back trajectories obtained for the aerosol samples collected during our cruises (see Figure S2). Aerosol samples of the SAfr-BB type had easterly arrivals at 1000 m, but southeasterly arrivals at the lower levels. Thus any day at points 3a2–4d which had 1000 m trajectories with a bearing < 123° to true north and an angle between the 50 m and 1000 m trajectories of 15°–38° was classified as SAfr-BB, provided it occurred during the biomass burning season (see Section 2.2). Then any other days with trajectories that touched southern Africa were classified as SAfr, as were all days for points 3b, 4a, 4b, 4c and 4d in which trajectories did not touch land, but which originated from close to land, defined as the area to the east of a line drawn between each point and Cape Agulhas (Lat 34.83°S, Lon 20.00°E). Back trajectories from point 3a2 were considered SAfr if they fell into the area east of this line and the differences between the bearings of the 3 and 5 day back trajectories were less than 10°. This was done to separate out any SAtl-Rem back trajectories which curved toward the southwest after 3 days. All other days for these points for which trajectories did not touch land were classified as SAtl-Rem. The total number of days that each air mass type occurred at each point was then aggregated, and these results are expressed as a percentage of the total period in Table A1.

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