

We follow the methodology of Kaufman et al. (2005), with several modifications. First, in addition to the zonal divergence of dust transport, we consider meridional divergence and temporal changes in dust mass:

$$D = -\nabla \cdot (M\mathbf{v}) - \frac{\partial M}{\partial t} \quad (1)$$

Here  $D$  represents bulk surface deposition,  $M$  is the mass concentration (total mass of dust in the atmosphere per unit area), and  $\mathbf{v}$  is the horizontal component of the wind at the level of the dust. According to (1), deposition is driven by the combination of dust transport convergence (first term on the right) and decreases in dust mass with time (second term on the right). The  $\partial M/\partial t$  term implicitly accounts for wet deposition. For example, if the wind speed and divergence are both zero but there is a decrease in dust mass with time due to rainfall, (1) will give positive values of deposition. Deposition is calculated from (1) at each  $1^\circ \times 1^\circ$  MODIS grid point using daily averages of  $\tau_{dust}$  and NCEP/NCAR reanalysis winds. The motivation for including the meridional divergence and time derivative in (1) is that deposition is an episodic event that is often associated with strong meridional transport (Fig. 1), and there are significant annual mean and seasonal variations of meridional winds that will affect the dust transport and deposition.

We found a fraction of fine aerosol for pure dust ( $f_{du}$ ) of 0.3 instead of 0.5 reported by Kaufman et al. (2005), based on the MODIS fine mode fraction averaged between  $12^\circ\text{N}$ – $20^\circ\text{N}$  during July–August for 2000–2012. The difference may result from the shorter time period or different region considered by Kaufman et al. (2005) compared to the present analysis. Using  $f_{du} = 0.3$  instead of  $f_{du} = 0.5$  decreases the dust optical thickness ( $\tau_{du}$ ) by one third.

Kaufman et al. (2005) set  $M/\tau_{du} = 2.7 \text{ g m}^{-2}$  based on the results of Tanré et al. (2001), Ginoux et al. (2001), Haywood et al. (2003), Maring et al. (2003), and Fan et al. (2004). However, other studies have resulted in  $M/\tau_{du}$  values ranging from 1.0 to 3.6 (Carlson 1979, Dulac et al. 1992, Moulin et al. 1997, Chin et al. 2002, Schepanski et al. 2009). We therefore use the median value reported by these studies, which is  $2.1 \text{ g m}^{-2}$ .

As dust is transported westward from Africa, larger particles settle more rapidly than small particles. We therefore allow  $M/\tau_{du}$  (and  $M$ , assuming that the light extinction efficiency is constant with particle radius) to decrease linearly by 10% from  $16^\circ\text{W}$  to  $25^\circ\text{W}$  and by an additional 15% between  $25^\circ\text{W}$  and  $60^\circ\text{W}$ . Here we follow the observational results of Maring et al. (2003) and assume that a higher fraction of the largest particles are deposited in the eastern basin.

Recent studies using CALIPSO show a strong seasonality in the average height of the dust layer (Generoso et al. 2008, Tsamalis et al. 2013, Wang et al., JAMC, 2014 submitted). Following their results, we allow the pressure levels at which  $\mathbf{v}$  is calculated in (1) to vary seasonally. During June–September a pressure of 700 hPa is used. During October–February a pressure of 900 hPa is used.

In regions of persistent cloud cover such as the ITCZ, there are about one tenth

as many daily estimates of deposition using (1) compared to the number of estimates using only the zonal divergence term (Fig. 2a,b). To obtain annual mean and seasonal deposition estimates that are more reliable, we therefore first calculate zonal convergence of dust mass concentration at each daily grid point. We then calculate deposition according to (1) and compare to simultaneous and co-located values of deposition from zonal convergence only. At each grid point, a linear regression is performed using all days when both estimates are available. The regression is done separately for the periods of October–May and June–September since the wind heights used in (1) are different. There is a strong linear dependence between zonal deposition and total deposition at most locations (Fig. 3).

The regression coefficients are used to scale the zonal convergence values in order to account for the meridional convergence and  $\partial M/\partial t$  terms. The justification is that zonal convergence has been shown to perform very well in the 10°N–25°N band (Kaufman et al. 2005, Das et al. 2013). On the other hand, meridional convergence and  $\partial M/\partial t$  are anticipated to be more important between the equator and 10°N and in the eastern basin, where lower level winds have a stronger southward component and removal of dust by precipitation is expected to become more important. Indeed, the mean correction to the zonal convergence estimates of deposition consists mainly of a meridional dipole that is strongest in the eastern basin, with positive values between about 5°N–12°N and negative values between 12°N–20°N (Fig. 2c), consistent with southward advection and convergence of dust. There is also a basin-wide increase in deposition of  $0.1 \mu\text{g m}^{-2} \text{s}^{-1}$ , equivalent to 43 Tg per year over the tropical North Atlantic Ocean (0°–25°N), when the meridional convergence and time terms are included. When only the zonal divergence is considered, our results agree with those of Kaufman et al. (2005), who estimate annual dust deposition to the tropical Atlantic Ocean of 144 Tg.

In order to separate wet deposition from dry deposition, we use daily measurements of rain rate from the TRMM satellite. Missing values are filled with the daily climatological rain rate (1998–2012) at that grid point. For a given MODIS grid point and day, if rainfall exceeds the threshold of  $0.02 \text{ mm hr}^{-1}$  (0.5 mm for the day), then the deposition on that day at that grid point is classified as wet. This is likely an upper bound on wet deposition based on the assumption that all deposition on a given day with rainfall is wet deposition, even if it was not raining for the entire day.

Independent estimates of dry deposition are obtained from the dust accumulation biases in the shortwave radiation records from several PIRATA moorings in the tropical North Atlantic (Foltz et al. 2013). The negative shortwave biases caused by dust buildup are first converted to a dust aerosol optical depth following the methodology of Evan and Mukhopadhyay (2010). Dust mass concentration is then calculated from the resultant  $\tau_{du}$  values following Kaufman et al. (2005) with the modifications described above. The resultant daily time series of dust buildup ( $M_{buoy}$ ) are interrupted by rainfall, which rinses the radiometer domes, and annual mooring servicing cruises, during which the radiometers with dust buildup are swapped out for new ones. We allow either of these occurrences to "reset" the  $M_{buoy}$  record to zero, following Foltz et

al. (2013). Fortunately, many of the servicing cruises have taken place during boreal summer and fall, after the rainy season has begun in the tropical North Atlantic and the radiometers have started to be rinsed. At most mooring locations there are therefore several months with continuous records of dust buildup, generally from November–January, after the rainy season has ended, until the start of the next rainy season in June–August.

The daily time series of  $M_{buoy}$  from each mooring is first smoothed with a 7-day running mean filter. To calculate annual mean deposition, the value of the smoothed  $M_{buoy}$  at the start of each calendar year is subtracted from the maximum value of the smoothed  $M_{buoy}$  within the same calendar year. This gives the maximum dust buildup (in  $\text{g m}^{-2}$ ) for a given calendar year. The maximum dust buildup is divided by the length of time during which the buildup occurred in order to estimate the rate of deposition onto the radiometer. These values can then be compared to the estimates based on (1). To estimate seasonal deposition from the moorings, a similar methodology is used, except the start days of December 1, March 1, June 1, and September 1 are used for each three-month season (DJF, MAM, JJA, SON, respectively). From these annual and seasonal estimates of deposition at each mooring location for each year, we calculate mean annual deposition, seasonal deposition, and interannual variability.

The dry dust deposition records from the PIRATA moorings are expected to underestimate the true dry deposition significantly, since it is anticipated that a large portion of the dust that falls onto a radiometer fails to stick to the instrument’s curved dome. However, it is found that the qualitative deposition estimates from the moorings are useful for validating the spatial and interannual variations of deposition obtained using (1).

The annual mean  $\tau_{dust}$  shows a maximum of 0.4 centered at about  $15^\circ\text{N}$  off the coast of Africa and a decrease westward to 0.1 at the entrance to the Caribbean (Fig. 4a). There is also a southward shift of about  $5^\circ$  in the latitude of maximum  $\tau_{dust}$  across the basin, consistent with the southward component of the trade winds and the concentration of dust lower in the atmosphere during boreal winter and spring. Annual mean dust deposition shows a similar southward shift in its maximum, from about  $15^\circ\text{N}$  off the coast of Africa to  $10^\circ\text{N}$  at  $50^\circ\text{W}$  (Fig. 4b). Dust deposition generally decreases westward, consistent with  $\tau_{dust}$ , but the decrease is less pronounced, especially  $30^\circ\text{W}$  and  $50^\circ\text{W}$ . There is a sharp meridional gradient of deposition at the approximate latitude of the center of the ITCZ ( $8^\circ\text{N}$  at  $20^\circ\text{W}$ ,  $5^\circ\text{N}$  at  $50^\circ\text{W}$ ), consistent with weaker winds and higher rainfall limiting dust transport and deposition.

There is a band of slightly negative values of deposition at  $20^\circ\text{W}$  between  $12^\circ\text{N}$  and  $20^\circ\text{N}$  that is caused by southward winds in the presence of southward-increasing  $\tau_{dust}$ , resulting in meridional divergence of dust mass (Fig. 4b). This may be caused by our use of an oversimplified and meridionally uniform dust height, whereas in reality the dust is expected to increase in height with latitude, resulting in a weaker southward component of transport. There is a larger region of negative deposition in the eastern basin between  $0^\circ$ – $5^\circ\text{N}$  that is also due to horizontal divergence of dust mass. Our basinwide estimates of dust deposition are therefore likely a lower bound on the true

deposition since they include these nonphysical negative values.

Deposition estimates from the PIRATA moorings are generally about a factor of ten lower than the satellite estimates (Fig. 4b). This is not surprising, since the mooring estimates generally cover only the first half of each year, are comprised entirely of dry deposition, and are based on the amount of dust that sticks to a curved dome. Nevertheless, the spatial pattern of deposition from the moorings agrees reasonably well with that from the satellite analysis. Both show the highest values in the eastern basin south of 20°N and lower values to the north and west. The mooring estimates also indicate that the highest rates occur along a northeast-southwest oriented line, consistent with the satellite estimates (Fig. 4b). The similar spatial patterns between the mooring dry deposition and satellite bulk deposition suggest that dry deposition may dominate in the eastern and central tropical North Atlantic, consistent with the satellite-based results (Figs. 4c, 5).

Wet deposition reaches a maximum in the western basin between 5°N–15°N, where mean rainfall is high ( $>5$  cm  $\text{mo}^{-1}$ ) and bulk deposition rates are high ( $>1$   $\mu\text{g m}^{-2} \text{s}^{-1}$ ) (Fig. 4c). Wet deposition accounts for as much as 40–50% of bulk deposition in the western ITCZ and as much as 80% in the extreme northwestern tropical Atlantic, consistent with the analysis of Prospero et al. (2010) for sites in Florida (Fig. 5). Wet deposition is close to zero in the northeastern tropical Atlantic, where rainfall is very infrequent.

Deposition rates in the 10°N–25°N band are positively correlated with  $\tau_{dust}$  averaged in the region (Fig. 6a). Most of the deposition in this region occurs in boreal summer, when transporting winds are westward and the maximum  $\tau_{dust}$  is in the eastern basin (Fig. 6b). In the 0°–10°N band deposition is positively correlated with area-averaged  $\tau_{dust}$  in the west, where winds have a stronger easterly component and  $\tau_{dust}$  decreases downwind, and negatively correlated in the east, where winds have a stronger meridional component and  $\tau_{dust}$  tends to increase downwind (Fig. 6c,d). Overall, these results are consistent with previous studies that have emphasized the strong relationship between  $\tau_{dust}$  and deposition rate (Das et al. 2013).

There are pronounced seasonal variations in  $\tau_{dust}$  and winds in the tropical Atlantic, which lead to a pronounced seasonal cycle in deposition. The mean July  $\tau_{dust}$  from MODIS shows a maximum of about 0.8 at 20°N directly west of Africa. The  $\tau_{dust}$  decreases to less than 0.3 at 60°W and the maximum shifts southward to 15°N (Fig. 7a). The zonal distribution of  $\tau_{dust}$  is consistent with transport by the 700 hPa winds, which are predominantly westward throughout the tropical North Atlantic, and a slight southward transport driven by the northeasterly trade winds. The highest values of  $\tau_{dust}$  are located to the north of the band of heavy rainfall associated with the ITCZ. In April  $\tau_{dust}$  peaks at about 0.5 at 10°N, 20°W and decreases to less than 0.3 at 6°N near the coast of South America (Fig. 7b). During April dust is lower in the atmosphere and as a result the meridional component of dust transport divergence becomes important. The combination of more southerly dust sources in April and southward dust transport results in a narrower meridional extent of the dust plume, with  $\tau_{dust}$  values greater than 0.2 confined to a 5° latitude band in the western basin

in April compared to  $10^\circ$  in July (Fig. 7).

Consistent with the seasonality in  $\tau_{dust}$ , the highest rates of dust deposition are concentrated between  $5^\circ\text{N}$ – $15^\circ\text{N}$  during boreal winter, spring, and fall, and between  $10^\circ\text{N}$ – $25^\circ\text{N}$  in the summer (Fig. 8). The highest deposition rates are concentrated mainly in the central and western basin during winter, spring, and fall, where wet deposition is also largest (Fig. 9). During the summer strong easterly winds at 700 hPa transport dust directly westward in the absence of significant rainfall, leading to the highest values of deposition in the east, where dry deposition dominates. The dry deposition estimates from the PIRATA moorings generally agree with the seasonality found in the MODIS data (Fig. 8). The highest values of deposition move northward from  $8^\circ\text{N}$ – $15^\circ\text{N}$  during December–May to  $15^\circ\text{N}$ – $20^\circ\text{N}$  during June–August (Fig. 8a-c). Lower values of deposition at the  $15^\circ\text{N}$  and  $20^\circ\text{N}$  moorings along  $38^\circ\text{W}$  in June–August compared to December–May may be caused by lower rates of accumulation of dust on the sensors when they are already coated. This also may explain why deposition at the  $12^\circ\text{N}$ ,  $23^\circ\text{W}$  mooring is so much higher in December–February ( $24 \mu\text{g m}^{-2} \text{s}^{-1}$ ) compared to March–May ( $3 \mu\text{g m}^{-2} \text{s}^{-1}$ ).

In the tropical North Atlantic Ocean ( $0^\circ$ – $25^\circ\text{N}$ ,  $15^\circ\text{W}$ – $60^\circ\text{W}$ ), we find a strong seasonal cycle in bulk deposition, with a pronounced peak of 118 Tg in boreal summer, which represents 62% of the annual deposition (Table 1). Bulk deposition has similar values in boreal spring and winter, and these seasons combined account for 35% of the annual deposition. Total deposition in the tropical Atlantic is found to be 191 Tg, which is 36% larger than found by Kaufman et al. (2005) using MODIS  $\tau_{dust}$ . The difference is due almost entirely to the inclusion of meridional and time terms in our calculation. Wet deposition accounts for only 17% of the annual total in the tropical Atlantic. This low amount is surprising, especially since the method used to calculate wet deposition is expected to yield an upper bound. It is possible that negative biases in bulk deposition in the ITCZ region, especially east of  $30^\circ\text{W}$  and possibly due to our neglect of meridional variations in the dust transport height, may lead to negative biases in wet deposition. Annual deposition to the Caribbean Sea is 37 Tg, about 17% as much as to the tropical Atlantic.

Pronounced interannual variations in  $\tau_{dust}$  have been observed in the tropical North Atlantic over the past 30 years (Lau and Kim 2007, Foltz and McPhaden 2008). To investigate the influence on dust deposition, we consider annual totals from MODIS and the PIRATA moorings. In the tropical North Atlantic Ocean ( $0^\circ$ – $25^\circ\text{N}$ ,  $15^\circ\text{W}$ – $60^\circ\text{W}$ ), deposition ranged from 120 Tg in 2001 to 210 Tg in 2007 (Fig. 10a). The  $\tau_{dust}$  and deposition time series are positively correlated at 0.5, which is significant at the 10% level. The positive correlation is due entirely to a correlation of 0.6 (significant at the 5% level) in the  $10^\circ\text{N}$ – $25^\circ\text{N}$  band, since the correlation in the  $0^\circ$ – $10^\circ\text{N}$  band is zero.

Though limited to only about half of the year, the deposition estimates from the PIRATA moorings offer an independent, though qualitative, assessment of interannual variability that can be compared to the MODIS estimates. In the northeastern tropical Atlantic ( $10^\circ\text{N}$ – $21^\circ\text{N}$ ,  $21^\circ\text{W}$ – $25^\circ\text{W}$ ), interannual variations of deposition and

$\tau_{dust}$  are similar to those averaged in the entire tropical North Atlantic (Fig. 10a,b), consistent with the high annual mean  $\tau_{dust}$  and deposition in this region (Fig. 4a). The deposition estimates from the 11.5°N, 23°W and 20.5°N, 23°W moorings during 2007–2012 generally agree with the area-averaged deposition from MODIS. Both moorings show maximum deposition in 2007–2008 and 2012, consistent with the MODIS values (Fig. 10b). Interestingly, 2010 had higher than average  $\tau_{dust}$ , but very low deposition. The difference was likely caused by anomalies in atmospheric circulation. In the central tropical North Atlantic (10°N–17°N, 36°W–40°W) the agreement between MODIS and mooring deposition is good between 2008–2012, when interannual variations correspond well to those of  $\tau_{dust}$  (Fig. 10c). However, MODIS deposition is poorly correlated with the mooring deposition and  $\tau_{dust}$  during 2001–2007. In contrast, the mooring deposition is highly correlated with  $\tau_{dust}$  during the whole period (0.8, significant at 1%). It is unclear whether the discrepancies are due to the limited measurement period (typically late boreal winter through early summer) and qualitative nature of the mooring estimates or from changes in atmospheric circulation, which would result in a lower correlation between MODIS deposition and  $\tau_{dust}$ .