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Kev Points:

- Greenland contributed
 0.74 ± 0.14 mm/yr to global mean sea level between 2011 and 2014
- Small-scale mass deficits have made a relatively large contribution to recent ice loss
- High-resolution radar altimetry can map Greenland mass balance at fine spatial and temporal resolution

Supporting Information:

- · Supporting Information S1
- Data Set S1

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A high-resolution record of Greenland mass balance

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Abstract We map recent Greenland Ice Sheet elevation change at high spatial (5 km) and temporal (monthly) resolution using CryoSat-2 altimetry. After correcting for the impact of changing snowpack properties associated with unprecedented surface melting in 2012, we find good agreement (3 cm/yr bias) with airborne measurements. With the aid of regional climate and firn modeling, we compute high spatial and temporal resolution records of Greenland mass evolution, which correlate (R = 0.96) with monthly satellite gravimetry and reveal glacier dynamic imbalance. During 2011–2014, Greenland mass loss averaged 269 ± 51 Gt/yr. Atmospherically driven losses were widespread, with surface melt variability driving large fluctuations in the annual mass deficit. Terminus regions of five dynamically thinning glaciers, which constitute less than 1% of Greenland's area, contributed more than 12% of the net ice loss. This high-resolution record demonstrates that mass deficits extending over small spatial and temporal scales have made a relatively large contribution to recent ice sheet imbalance.

1. Introduction

Since the early 1990s, mass loss from the Greenland Ice Sheet has contributed approximately 10% of the observed global mean sea level rise [Vaughan et al., 2013]. During this period, the ice imbalance has increased with time [Rignot et al., 2011; Shepherd et al., 2012], as warmer atmospheric conditions have prevailed [Hanna et al., 2008; Fettweis et al., 2013a, 2013b] and many marine-terminating glaciers have accelerated [Joughin et al., 2010; Moon et al., 2012; Enderlin et al., 2014]. Between 2000 and 2008, ice loss was due, in almost equal measures, to decreased surface mass balance (SMB) and increased ice discharge [van den Broeke et al., 2009]. Since then, several exceptionally warm summers [Hanna et al., 2012] have produced episodes of widespread surface melt [Nghiem et al., 2012; Fettweis et al., 2013b]. These conditions, which have been linked to the advection of warmer southerly air over the ice sheet [Fettweis et al., 2013a; Tedesco et al., 2013], have further increased annual ice losses from atmospheric melting and subsequent runoff [Schrama et al., 2014]. As a result, between 2009 and 2012, surface mass balance increased its contribution to 70% of the total ice sheet imbalance [Enderlin et al., 2014].

Recent monitoring of the Greenland Ice Sheet has illustrated the high temporal and spatial variability in ice loss [Rignot et al., 2011; Moon et al., 2012; Csatho et al., 2014; Enderlin et al., 2014; Schrama et al., 2014]. In 2012, annual mass losses of approximately 500 Gt were recorded [Tedesco et al., 2013], representing the largest deficit of the observational era, yet these were followed in 2013 by a return to a state of near balance [Khan et al., 2015]. Similarly, there has been substantial interregional and intraregional variability in glacier dynamics [Moon et al., 2012], as the response of individual systems to regional climatic forcing can be modulated by varying glacier geometry and setting. The high spatial and temporal variability in Greenland glacier behavior complicates extrapolation of recent observations forward in time and reinforces the need to develop process-based projections of ice sheet evolution. This requires an understanding of the drivers and timescales of ice sheet change, which in turn demands observations of ice sheet mass evolution sampled with sufficiently high spatial and temporal frequency to resolve this variability. Previous studies have demonstrated that CryoSat-2 radar altimetry can successfully resolve changes in ice sheet elevation and volume [Helm et al., 2014]. Here we use CryoSat-2 radar altimetry to produce high spatial (5 km) and temporal (monthly) records of Greenland Ice Sheet mass evolution, and use these measurements to resolve the detailed pattern of ice loss between 2011 and 2014.

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2. Data and Methods

We used CryoSat-2 radar altimetry to compute both linear rates of Greenland mass balance and monthly ice mass evolution between 1 January 2011 and 31 December 2014. We computed rates of elevation change within 5×5 km grid cells using a model fit method [Smith et al., 2009; Flament and Rémy, 2012; McMillan et al., 2014a] to partition the elevation fluctuations recorded within each grid cell, according to the contributions from topography, temporal changes in elevation, and varying radar backscatter. The model was adapted from previous studies [McMillan et al., 2014a, 2014b] to account for the impact of changing snowpack characteristics on the backscattered echo. Varying snowpack liquid water content, density, and roughness can alter the depth distribution of the backscattered energy and impact upon radar altimeter elevation measurements [Scott et al., 2006; Gray et al., 2015]. Although we expect seasonal cycles in backscatter depth to have only a limited impact on longer-term elevation trends, more occasional, episodic changes in scattering characteristics have the potential to introduce larger artifacts into the elevation record. Such an effect occurred over the Greenland Ice Sheet interior during the summer of 2012 [Nilsson et al., 2015] when approximately one third of the ice sheet experienced melt for the first time in a decade [Tedesco et al., 2013].

To investigate the impact of the 2012 melt event on the radar altimeter measurements, we applied a numerical deconvolution procedure [Arthern et al., 2001] to radar echoes acquired across the ice sheet interior during this time (see Text S2 in the supporting information). This enabled us to estimate the distribution of backscattered power as a function of depth within the snowpack, by removing the contribution from reflections within the radar beam footprint that were beyond the point of closest approach. This analysis suggested that after the interior of the ice sheet experienced melt conditions in July 2012, there was a widespread transition from volume to surface scattering, a sharp increase in the extinction coefficient, and an abrupt increase in the elevation recorded by the altimeter (Text S2), consistent with the interpretation of Nilsson et al. [2015]. We therefore adapted our model fit approach to accommodate this abrupt shift in dominant scattering horizon across the ice sheet interior during July 2012, using a step change function to model the recorded elevation change associated with this event (Text S3). To evaluate our approach, we compared our results to 8149 estimates of elevation change derived from colocated IceBridge airborne altimetry measurements [Krabill, 2014], acquired between March 2009 and May 2014 (Figure 1). The mean difference in the elevation change rates (CryoSat-2-IceBridge) was reduced from 9 cm/yr to 3 cm/yr by accounting for the 2012 melt event, and the standard deviation of the resulting differences was 65 cm/yr.

We then converted the altimeter-derived rates of change to estimates of Greenland mass balance. For this conversion two approaches are well established. Either a density model is used, which accounts for known dynamic and SMB processes [Davis et al., 2005; Thomas et al., 2006; Wingham et al., 2006; Shepherd and Wingham, 2007; Sørensen et al., 2011] or simulations of firn column thickness change are removed from the observed signal to estimate the underlying ice imbalance [Li and Zwally, 2011; Zwally et al., 2011]. With the latter approach, the modeled firn mass changes are then added back to the ice mass changes, to estimate the total mass balance. In this study, we choose to use the former method because (1) it avoids errors in the observed and modeled elevation rates being interpreted as widespread dynamic imbalance across the interior of the ice sheet and (2) it eliminates the need to correct for the 50%-70% of the observed signal that is due to surface processes [van den Broeke et al., 2009; Enderlin et al., 2014] in order to estimate the residual changes due to dynamic imbalance. Further comparison of the two methods is provided in section 3 and in Text S6 of the supporting information.

In common with other recent altimetry estimates of Greenland mass balance [Sørensen et al., 2011], prior to the volume-to-mass conversion, we applied a firn compaction correction. This step was designed to account for mass-conserving fluctuations in the rate of firn compaction, as any deviation of this rate away from steady state conditions will cause an elevation change that is not associated with a change in mass [Zwally et al., 2005]. Specifically, we removed the component of the observed rate of elevation change that was due to firn compaction anomalies during our study period, using simulations of firn compaction from the Institute for Marine and Atmospheric Research Utrecht Firn Densification Model (IMAU-FDM v1.0) [Ligtenberg et al., 2011; Kuipers Munneke et al., 2015]. We emphasize that this procedure is designed to correct solely for mass-conserving processes, in order to isolate signals related to changing ice sheet mass, and so we only remove the simulated elevation change due to firn compaction anomalies. This step does not remove the total firn column elevation change, which also includes surface mass deposition and removal. IMAU-FDM

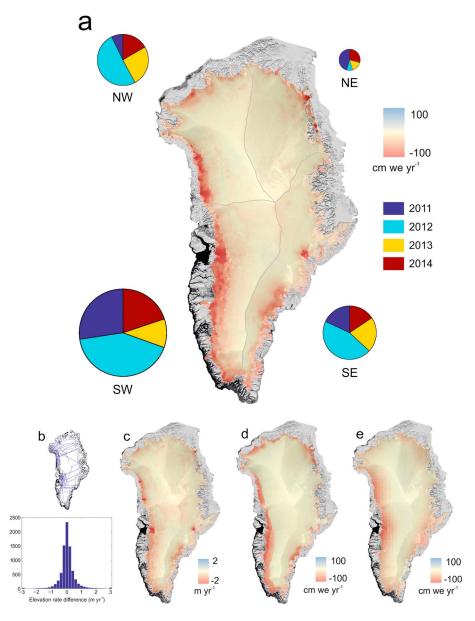


Figure 1. Greenland mass and elevation change. (a) Rate of mass change between January 2011 and December 2014 from CryoSat-2 radar altimetry and firn modeling. For each of the southwest (SW), southeast (SE), northeast (NE), and northwest (NW) sectors, the color wheel indicates the proportion of mass lost in each year, with the radius scaled according to the magnitude of the total losses. The boundaries between the four sectors are shown in gray. (b) The distribution of IceBridge airborne altimetry tracks used for validation and a histogram of the differences (CryoSat-2 minus IceBridge) in the recorded rates of elevation change. (c) Rate of elevation change between 2011 and 2014 from CryoSat-2 radar altimetry. (d) Simulated 2011–2014 rate of mass change due to surface processes from RACMO2.3 regional climate modeling. (e) Rate of mass change between 2011 and 2014 from GRACE gravimetry. The CryoSat-2 maps have been smoothed with a 25 by 25 km median filter and the background image is the MODIS mosaic of Greenland [Haran et al., 2013] overlaid with a shaded relief of ice sheet elevation.

simulates the temporal evolution of firn compaction, meltwater percolation and refreezing, and temperature in a vertical, 1-D column of firn and ice. The model is forced at its upper boundary by RACMO2.3, the most recent version of the RACMO regional climate model [Noël et al., 2015]. Both models operate at 11×11 km spatial resolution.

To convert the resulting altimeter rates of change to mass, we constructed a density model that accounted for both surface and dynamic processes. In regions where high rates of elevation change and ice flow suggested a state of dynamic imbalance, we used an ice density of 917 kg m $^{-3}$ (see Text S8). Elsewhere, detected elevation

changes were assumed to be driven by SMB processes, and we used an ice density within the ablation zone and the density of the IMAU-FDM firn layers gained or lost across the remaining areas. We then estimated the total ice sheet mass balance by summing the individual grid cell contributions and filled the 11% of grid cells where no elevation rate was retrieved, using a model based on elevation, latitude, and velocity change (see Text S4). The total mass balance uncertainty was computed by estimating spatially correlated and uncorrelated uncertainty in the measured surface elevation change, associated with changing snowpack characteristics and measurement imprecision, respectively. These were combined with the uncertainties arising from the unobserved areas, the density model, and the magnitude of the Glacial Isostatic Adjustment (GIA) and elastic bedrock uplift signals (see Text S9).

We also used the CryoSat-2 measurements to compute time-varying mass evolution at monthly intervals, by using our model fit solution to isolate the change, with time, of elevation anomalies within each grid cell. These anomalies were averaged over monthly intervals, corrected for the influence of firn compaction and converted to mass using monthly densities from the IMAU-FDM, supplemented with our dynamic imbalance mask. Monthly densities were used in order to account for seasonal variations in the density of surface mass change. At each monthly time step, we then used a bilinear interpolation to fill unobserved regions between satellite ground tracks, integrated spatially to produce an ice sheet mass anomaly and scaled our results in accordance with the proportion of the mass balance field sampled. The latter step was used to account for the bias introduced by preferentially sampling high latitude and inland regions (see Text S6).

To evaluate our estimates of ice sheet mass balance, we used independent observations from the Gravity Recovery and Climate Experiment (GRACE) satellite and modeling results from the RACMO2.3 regional climate model [Ettema et al., 2009; Noël et al., 2015]. Monthly GRACE solutions from the University of Texas Center for Space Research, with maximum spherical harmonic degree 96, were used to estimate ice mass changes following the regional integration approach [Swenson and Wahr, 2002; Horwath and Dietrich, 2009]. The integration kernel was tailored to minimize the combined effect of GRACE errors and leakage errors, and specifically the sensitivity to mass change in the Canadian Arctic, although some impact from these glaciers inevitably remains. The estimate necessarily includes mass changes of peripheral glaciers, because the limited spatial resolution of GRACE prevents their separation without external information. We corrected for gravity field changes due to GIA using modeling results by A et al. [2013], based on the ICE-5G glaciation history [Peltier, 2004]. We estimated the uncertainty on GRACE Greenland mass trends based upon simulations of the leakage error and an analysis of the GIA correction uncertainty by Velicogna and Wahr [2013].

RACMO2.3 simulations [Noël et al., 2015] were used to estimate the rates of mass change driven by surface processes during our study period. The rate of mass change was estimated from a linear fit to the modeled cumulative surface mass balance anomalies, relative to the 1960-1979 average. The associated uncertainty accounted for both systematic and time-varying model uncertainties, together with the precision of the linear fit (see Text S10).

3. Results and Discussion

3.1. Regional Variability in Ice Loss

Between 1 January 2011 and 31 December 2014 we estimate that the Greenland Ice Sheet lost mass at a rate of 269 ± 51 Gt/yr. The high spatial resolution and comprehensive coverage provided by CryoSat-2 provides a detailed map of ice sheet mass balance, showing widespread ice loss at lower elevations, particularly along the western margin, with the highest rates of change located across several distinct glacier systems (Figure 1). We find that in recent years the southwestern sector has been the dominant source of mass loss, contributing 41% of the total 4 year deficit. In contrast, the northeast contributed only 10% of all losses, with the remainder split between the southeast (25%) and northwest (24%) sectors. Our observations indicate that although regional mass imbalance has been most pronounced at lower latitudes, ice has also been lost from much of the northeast and northwest margins. These findings, which are supported by the RACMO2.3 simulations (Figure 1), provide observational evidence that atmospheric conditions are affecting the far north of the ice sheet.

The pattern of regional mass balance resolved by CryoSat-2 is broadly similar to that derived from satellite gravimetry (Figure 1). Using GRACE, we find a total ice sheet mass balance over the same period of -279 \pm 22 Gt/yr, although it is important to note that the GRACE estimate also integrates the mass change of

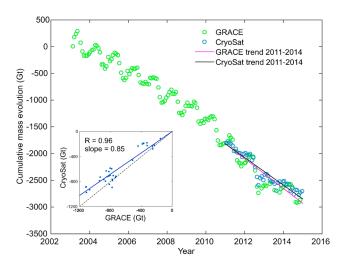


Figure 2. Greenland mass evolution. Monthly evolution in ice sheet mass since 2003 from GRACE gravimetry (green) and since 2011 from CryoSat-2 altimetry and firn modeling (blue). The CryoSat-2 time series has been referenced to the GRACE data at the start of 2011. The inset shows the correspondence between the GRACE and CryoSat-2 monthly estimates of mass evolution since 2011 (solid blue dots), together with a linear regression (solid blue line), the regression slope, and the Pearson correlation coefficient, *R*. The dashed line indicates equivalence, although the GRACE results include, additionally, mass changes of peripheral ice caps and unglaciated regions.

peripheral ice caps, which in recent years have contributed a further 30-40 Gt/yr of ice loss [Bolch et al., 2013]. In comparison to GRACE, the uncertainty associated with our CryoSat-2 estimate is relatively large, although the finer spatial detail is able to resolve mass balance at the scale of individual glacier catchments. The higher CryoSat-2 uncertainty largely reflects the difficulty in assessing the influence of spatially correlated changes in snowpack properties. In this study, we have estimated this uncertainty based upon the impact of the extreme 2012 melt event (Text S9), which yields an uncertainty slightly above the 45 Gt/yr that would be expected given the mean difference between the rates of elevation change recorded by CryoSat-2 and IceBridge altimetry. Further work is needed to formally constrain this component of the uncertainty budget.

Over the same spatial and temporal domain as our CryoSat-2 observations, RACMO2.3 indicates that atmospheric processes produced an annual SMB deficit of -154 ± 65 Gt/yr, which is equivalent to 57% of the observed ice sheet mass balance. The difference of -115 ± 83 Gt/yr, which represents the mass loss due to other factors, is close to an independent estimate of the 2009–2012 dynamic loss of 122 Gt/yr [Enderlin et al., 2014], although our associated uncertainty is large when integrated over the whole ice sheet, reflecting the challenges of partitioning the observed signals across the entire ice sheet interior. We investigate the differences between the two data sets and their utility for detecting dynamic imbalance at the more localized scale of individual glaciers catchments, in more detail below.

3.2. Temporal Variability in Ice Loss

Our monthly estimates of ice sheet mass evolution resolve the temporal variability associated with seasonal and interannual forcing, and also enable an assessment of the consistency between the CryoSat-2 and GRACE records (Figure 2). Both techniques resolve large variations in annual ice loss. The CryoSat-2 measurements show that losses during the exceptionally warm summer of 2012 produced an annual ice deficit of 439 ±62 Gt, which dominated the total 4 year period. In contrast, 2013 saw only moderate losses of 116±65 Gt, which were approximately half the 2000-2011 mean [Shepherd et al., 2012]. Comparing the monthly CryoSat-2 and GRACE estimates, we find close agreement (Pearson correlation coefficient, R, of 0.96) between the two independent approaches, albeit over their different spatial domains. This statistic reflects, in part, the consistency with which both methods resolve the long-term trend in mass loss. There is, however, also a relatively good agreement (R = 0.78) in the seasonal and interannual variability, once the long-term trend is removed. The GRACE observations do tend to exhibit slightly larger amplitude seasonal cycles, and the 2012 deficit recorded by CryoSat-2 is lower than a previous GRACE estimate [Tedesco et al., 2013]. This may be because of the incomplete sampling by CryoSat-2 of the full mass field at each monthly epoch, possible dampening effects caused by the CryoSat-2 backscatter correction, the limited sensitivity of radar altimetry to seasonal cycles in dry snow accumulation or the integration by GRACE of signals arising from peripheral ice caps and unglaciated regions.

Our record shows that during the survey period, annual ice losses varied by several hundred gigatons. Analyzing the RACMO2.3 surface mass fluxes and comparing these with estimates of ice discharge from *Enderlin et al.* [2014] indicates that the dominant source of this variability was fluctuations in meltwater runoff.

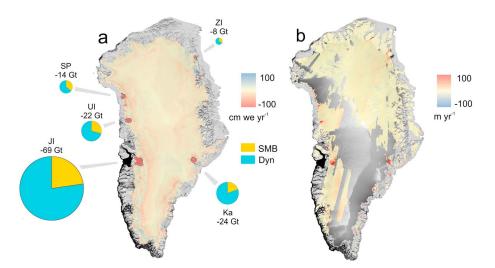


Figure 3. Greenland dynamic mass balance, 2011–2014. (a) Difference between Cryosat-2 and RACMO2.3 mass balance. (b) Ice velocity change between 2000–2001 and 2008–2009 mapped by imaging Synthetic Aperture Radar [Joughin et al., 2010]. Several marine-terminating glaciers exhibiting signs of dynamic ice loss are apparent; Kangerdlugssuag (Ka), Jacobshavn Isbræ (JI), Upernavik Isstrøm (UI), Steenstrup (SP), and Zachariae Isstrøm (ZI); all of which have undergone velocity change during the preceding decade. For each of these glaciers, the total 4 year mass loss from the rapidly thinning terminus regions, bounded by the gray lines, is shown. The color wheels indicate the partitioning of mass losses within the bounded regions, according to surface mass and dynamic processes, with the radius of each wheel scaled according to the magnitude of the total losses. The background image is the MODIS mosaic of Greenland [Haran et al., 2013] overlaid with a shaded relief of ice sheet elevation.

Whereas year-to-year differences in regionally integrated solid ice discharge typically do not exceed 20 Gt/yr (~5% of the mean) and interannual variability in snowfall is also relatively modest (1991–2014 standard deviation of 61 Gt/yr or 9% of the mean), meltwater runoff has exhibited much larger variability. Between 1991 and 2014, the standard deviation of annual runoff was 102 Gt/yr or 28% of the mean. In recent years, contrasting atmospheric conditions have driven even greater variability, with annual runoff of 625 Gt/yr and 298 Gt/yr in 2012 and 2013, respectively. In 2012, intense summer melting was driven by persistent high pressure, associated with a strong negative phase of the North Atlantic Oscillation (NAO) and a high Greenland Blocking Index [Hanna et al., 2014]. In contrast, 2013 saw low-pressure and low-temperature conditions, coinciding with the most positive summertime NAO recorded in the past 20 years. Our record demonstrates the impact that these changing atmospheric conditions have had upon year-to-year variability in mass loss and more specifically the potential for intense melt events lasting only a few months to make large contributions to multiyear mass balance.

3.3. Mass Loss Due To Dynamic Processes

Differencing the CryoSat-2 and RACMO2.3 mass balance fields removed the simulated component due to atmospheric processes. This allowed us to investigate the discrepancies between the two data sets, due to model or observational errors, and to identify individual glacier systems where additional mass changes suggested dynamic imbalance (Figure 3). To evaluate these signals, we also computed recent changes in glacier flow using repeated ice velocity measurements from 2000-2001 and 2008-2009 [Joughin et al., 2010]. The CryoSat-2 and RAMCO2.3 data sets provide evidence of significant dynamic mass loss at five marineterminating glaciers; Kangerdlugssuag Glacier in the southeast; Jacobshavn, Upernavik Isstrøm, and Steenstrup Glaciers on the western margin, and Zachariae Isstrøm in the northeast (Figure 3a). Significant signals of dynamic imbalance are not apparent in the far southeast, which may relate to the incomplete sampling of these coastal regions. Comparing our mass loss and velocity measurements, we find that each of the five glaciers identified above has undergone a significant increase in velocity during the preceding decade (Figure 3b), suggesting that deficits in mass associated with these flow accelerations have continued throughout the duration of this study. With its interferometric capability, CryoSat-2 is able to sample 67% of the near-terminus areas of these glaciers where the dynamic imbalance is most pronounced. These regions, which together constitute only 0.9% of the total ice sheet area, have contributed more than 12%

of the total mass balance during our study period. In contrast, Storstrømmen in the northeast is the only glacier where we detect dynamically driven mass gain. This is likely to be an ongoing response to flow deceleration since the glacier surged between 1978 and 1984 [Mohr et al., 1998].

Our results demonstrate the capability of interferometric altimetry and regional climate modeling to monitor ice dynamic imbalance at the catchment scale. At the highest elevations, however, CryoSat-2 tends to show more negative mass change than RACMO2.3, whereas across some land-terminating sectors, particularly in the southwest, the reverse is found. Although multidecadal deceleration in ice velocity has been observed across parts of the ablation zone of western Greenland [van de Wal et al., 2008; Tedstone et al., 2015], direct evidence of changes in strain rate and associated dynamic thickening, is still lacking. Rather, we believe that these observed differences within the interior are indicative of the respective uncertainties associated with the two data sets. These may arise from uncompensated altimeter signals related to changing snowpack characteristics, the initialization profile used by the firn densification model, or the inability of RACMO2.3 to completely reproduce the temporal variability in the cumulative surface mass balance anomaly. Together, these differences reflect the current challenges associated with mapping and modeling Greenland mass balance fields at high resolution and motivate our decision to employ a density model-based approach within this study (Text S6). Nonetheless, other altimeter-based methods have been advocated within the community, whereby the difference between the total (observed) and SMB (modeled) components of mass change are used to estimate the underlying ice imbalance across the entire ice sheet. For comparison we therefore also compute mass change using this approach. This method yields an estimated mass balance of -230 Gt/yr, which is 39 Gt/yr more positive than our chosen approach, because it attributes all residual signals across slow-flowing regions to result from dynamic imbalance. Determining with confidence the extent of any dynamic changes across the slow-flowing interior remains, we believe, a challenge and a principal avenue for future research.

4. Conclusions

Using CryoSat-2 radar altimetry and the RACMO2.3 and IMAU-FDM models, we have computed Greenland Ice Sheet mass balance at high spatial and temporal resolution. Despite the challenges associated with radar wave interaction with a highly variable snowpack, we find a good level of agreement with both airborne altimetry and satellite gravimetry. Between January 2011 and December 2014 we estimate that the Greenland Ice Sheet lost an average of 269 ± 51 Gt/yr of snow and ice. The observed deficit indicates an annual contribution of 0.74 ± 0.14 mm/yr to global mean sea level, which is approximately double the 1992–2011 mean [Shepherd et al., 2012]. Since 2011, ice sheet mass balance has been highly variable in space and time. After the record deficit of 439 ± 62 Gt observed in 2012, which was driven by an exceptionally warm summer, subsequent ice losses have been more moderate. Combining altimetry and regional atmospheric model simulations, we identify five glacier systems which currently exhibit negative ice dynamic balance, all of which have undergone significant changes in velocity during the preceding decade. Our results suggest that future Synthetic Aperture Radar altimetry missions can contribute toward monitoring and understanding ongoing Greenland mass balance at the scale of individual glacier systems.

This work was supported by the UK Natural Environment Research Council. The CryoSat-2 satellite altimetry data are freely available from the European Space Agency (https://earth.esa.int/ web/guest/data-access) and the specific data used in this study are provided within the supporting information. The IceBridge airborne altimetry data are freely available from the National Snow and Ice Data Centre (https://nsidc.org/ data/icebridge/). The GRACE data are freely available from the Physical Oceanography Distributed Active Archive Center (PO.DAAC) at the Jet Propulsion Laboratory (http://podaac. jpl.nasa.gov/grace). The ice velocity data are freely available from the National Snow and Ice Data Centre (http://nsidc.org/data/docs/measures/ nsidc0478_joughin/). P.K.M., M.R.v.d.B., W.J.v.d.B., B.P.Y.N., and S.R.M.L. acknowledge financial support from the Polar Program of the Netherlands Organization for Scientific Research (NWO). We are grateful to three anonvmous reviewers and the editors, whose comments have significantly improved the manuscript.

Acknowledgments

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