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Strong constraints on aerosol-cloud interactions from volcanic eruptions

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45

46 **Summary (149 words of referenced text):**

47 The climate impact of aerosols is highly uncertain owing primarily to their poorly quantified
48 influence on cloud properties. During 2014-15, a fissure eruption in Holuhraun (Iceland)
49 emitted huge quantities of sulphur dioxide, resulting in significant reductions in liquid cloud
50 droplet size. Using satellite observations and detailed modelling, we estimate a global mean
51 radiative forcing from the resulting aerosol-induced cloud brightening for the time of the
52 eruption of around -0.2 W.m^{-2} . Changes in cloud amount or liquid water path are
53 undetectable, indicating that these aerosol-cloud indirect effects are modest. It supports the

54 idea that cloud systems are well buffered against aerosol changes as only impacts on cloud
55 effective radius appear relevant from a climate perspective, thus providing a strong constraint
56 on aerosol-cloud interactions. This result will reduce uncertainties in future climate
57 projections as we are able to reject the results from climate models with an excessive liquid
58 water path response.

59

60 **Main Text: (3103 words of referenced text, including concluding paragraph)**

61 **1. The 2014-15 eruption at Holuhraun (486 words of referenced text):**

62 Anthropogenic emissions that affect climate are not just confined to greenhouse gases.
63 Sulphur dioxide and other pollutants form atmospheric aerosols that can scatter and absorb
64 sunlight and can influence the properties of clouds, modulating the Earth-atmosphere energy
65 balance. Aerosols act as cloud condensation nuclei (CCN); an increase in CCN translates into
66 a higher number of smaller, more reflective cloud droplets that scatter more sunlight back to
67 space¹ (the ‘first’ indirect effect of aerosols). Smaller cloud droplets decrease the efficiency
68 of collision-coalescence processes that are pivotal in rain initiation, thus aerosol-influenced
69 clouds may retain more liquid water and extend coverage/lifetime^{2,3} (the ‘second’ or ‘cloud
70 lifetime’ indirect effect). Aerosols usually co-vary with key environmental variables making
71 it difficult to disentangle aerosol-cloud impacts from meteorological variability⁴⁻⁶.
72 Additionally, clouds themselves are complex transient systems subject to dynamical
73 feedbacks (e.g. cloud top entrainment/evaporation, invigoration of convection) which
74 influence cloud response⁷⁻¹². These aspects present great challenges in evaluating and
75 constraining aerosol-cloud interactions (ACI) in General Circulation Models (GCM)¹³⁻¹⁷,
76 with particular contentious debate surrounding the relative importance of these feedback
77 mechanisms.

78 Nonetheless, anthropogenic aerosol emissions are thought to cool the Earth via indirect
79 effects¹⁷, but the uncertainty ranges from -1.2 to -0.0 W.m⁻² (90% confidence interval) due to
80 *i*) a lack of characterization of the pre-industrial aerosol state^{15,18,19}, and *ii*) model parametric

81 and structural errors in representing cloud responses to aerosol changes^{16,18,20,21}. It is
82 estimated that uncertainty in the pre-industrial state can account for approximately 30% of
83 total ACI uncertainty^{18,21} while representation of chemistry-aerosol-cloud processes in
84 models is responsible for the remaining 70% uncertainty^{16,21}. Recently, a framework to break
85 down uncertainties in the causal chain from emission to radiative forcing showed that the
86 sources of uncertainty within different GCMs differ greatly¹⁶.

87 Volcanic eruptions provide invaluable natural experiments to investigate the role of large-
88 scale aerosol injection in the Earth system²²⁻²⁶. There have been several Icelandic volcanic
89 eruptions over recent years; Eyjafjallajökull erupted in 2010, Grímsvötn in 2011 and
90 Holuhraun in 2014-15. At its peak, the 2014-15 eruption at Holuhraun emitted ~120 kt of
91 sulphur dioxide (SO₂) per day into the atmosphere, a rate some four times higher than all 28
92 European Union member states or over a third of global emission rates. Iceland became in
93 effect a continental-scale pollution source of SO₂; SO₂ is readily oxidised via gas- and
94 aqueous-phase reactions, producing a massive aerosol plume in a near-pristine environment
95 where clouds should be most susceptible to aerosol concentrations^{16,18,27}.

96 We advance upon preliminary observational assessments of the impact of the 2014-15
97 eruption at Holuhraun^{28,29} through an extensive observational analysis that includes a
98 statistical evaluation of the significance of the observed spatial distribution of the cloud
99 perturbations to untangle the impacts of aerosol/meteorological impacts. We then assess the
100 simulation from a range of different climate models and assess the performance against
101 available observations. Finally, we show that observations of a volcanic plume (Mt. Kilauea,
102 Hawaii) in an entirely different meteorological regime exhibit similar overall impacts.

103

104 **2. Impact of the eruption on clouds (2140 - 20 = 2120 words of referenced text):**

105 Following the lifecycle of sulphur from emission, our initial analysis concentrates on the
106 coherence of SO₂ detected by the Infrared Atmospheric Sounding Interferometer (IASI)
107 sensor (Supplementary M1) and the HadGEM3 GCM that is constrained by observed

108 temperatures and winds (i.e. nudged, Supplementary M2). IASI retrievals use the discrete
109 spectral absorption structure of SO₂ to determine concentrations³⁰. Comparisons of IASI SO₂
110 observations from explosive volcanic eruptions against model simulations have proven
111 valuable in the past^{31,32}. The processing procedure for quantitative comparison between IASI
112 and HadGEM3 data uses only data that are spatially and temporally coherent (Supplementary
113 M3).

114 There is considerable uncertainty in the quantitative emission of SO₂ from the 2014-15
115 eruption at Holuhraun. A previous study²⁸ assumed a constant emission rate of 40
116 kt[SO₂]/day based on initial estimates of degassing. As our standard scenario (STAN) we use
117 an empirical relationship between degassed sulphur and TiO₂/FeO ratios and lava production
118 derived from Icelandic basaltic flood lava eruptions³³ which suggests significantly higher
119 emissions during the early phase of the eruption in September, but we also investigate a
120 simulation where a constant 40 ktSO₂/day is released (40KT scenario). The model
121 simulations and IASI retrievals of column SO₂ are shown in Figure 1 (40KT emission
122 scenario shown in Supplementary S1).

123

124 *****Insert Figure 1 here*****

125

126 The distribution and the magnitude of the column loading of SO₂ detected by IASI are similar
127 to those derived from HadGEM3, showing that the GCM nudging scheme and the assumed
128 altitude of the emissions in the STAN scenario (surface to 3 km) reproduces the week to
129 week spatial variability and magnitude of observed column SO₂ (SI-SO2_animation.mp4).

130 While the spatial distribution of sulphate aerosol optical depth (*AOD*) caused by the eruption
131 can be determined easily in the model (Supplementary Fig. S2.1), detection of the aerosol
132 plume over the north Atlantic in the MODIS data is hampered by the mutual exclusivity of
133 aerosol and cloud retrievals. The predominance of cloudy scenes makes accurate detection of
134 the aerosol plume in monthly-mean MODIS data extremely challenging (Supplementary S2).

135 Nonetheless, despite lacking observations of AOD , we can look for evidence of perturbations
136 caused by aerosols on cloud properties. We examine the perturbation to retrieved cloud top
137 droplet effective radius (r_{eff}) in September and October 2014 using collection 051 monthly
138 mean data from MODIS AQUA (MYD08, Supplementary M4) over the period 2002-2014.
139 MODIS AQUA data are not subject to the degradation in performance of the sensors at
140 visible wavelengths that has recently been documented for the MODIS TERRA³⁴ sensor
141 (Supplementary S3). We present a summary of the change in r_{eff} , Δr_{eff} , for October 2014
142 compared to the long term 2002-2013 mean in Figure 2a. A full analysis of the year-to-year
143 variability in Δr_{eff} is presented in Supplementary S4.

144

145 *****Insert Figure 2 here*****

146

147 There is clear evidence of a signal in Δr_{eff} in October (Figures 2a) and September
148 (Supplementary Fig. S5.1a). Pixels that are statistically significantly different from the 2002-
149 2013 climatological mean at 95% confidence occur over the entire breadth of the north
150 Atlantic. The spatial distribution of Δr_{eff} is governed by the prevailing wind conditions that
151 advect the volcanic plume and are quantitatively similar to those noted in Collection 006
152 MODIS data²⁹.

153 Figures 3a show the corresponding Δr_{eff} derived from the model in October (for September,
154 Supplementary Fig. S5.2a). The observations and modelling show obvious similarities in
155 spatial distribution. In addition to the spatial coherence in Δr_{eff} , the changes in the model
156 of $-1.21 \mu\text{m}$ (September) and $-0.68 \mu\text{m}$ (October) are within 30% of MODIS Δr_{eff} of $-0.98 \mu\text{m}$
157 (September) and $-0.97 \mu\text{m}$ (October) for the domain shown in Figure 2.

158

159 *****Insert Fig 3 here*****

160

161 There are similarities between the MODIS and HadGEM3 probability distribution functions
162 (Figures 2b and 3b) with a shift to smaller r_{eff} for the year of the eruption. Almost all high
163 values of r_{eff} (i.e. $r_{eff} > \sim 16 \mu\text{m}$ for MODIS and $r_{eff} > \sim 11 \mu\text{m}$ for HadGEM3) are absent in
164 2014 suggesting that clouds with high r_{eff} are entirely absent from the domain in both the
165 observations and the model. There are obvious discrepancies in the absolute magnitude of r_{eff}
166 between MODIS and HadGEM3. MODIS retrievals of r_{eff} from the MYD06 product in liquid
167 water cloud regimes have been shown to be significantly larger than those derived from other
168 satellite sensor products, mainly due to the algorithm's use of a different primary spectral
169 channel relative to other products^{35,36}. Nevertheless, Δr_{eff} is in encouraging agreement as this
170 quantity, along with changes in cloud liquid water path (LWP), needs to be accurately
171 represented if aerosol-cloud interactions are to be better quantified. As with r_{eff} , there are
172 similarities between the MODIS and HadGEM3 for ΔLWP (Figure 2c-d and Figure 3c-d),
173 however, evidence of a clear signal due to the volcano is neither observed or modelled.
174 Additionally, we also found that perturbations in the monthly mean cloud fraction from
175 MODIS are negligible, both in September and October as previously reported²⁹.

176 It is incumbent on any study attributing Δr_{eff} to volcanic emissions to prove the causality
177 beyond reasonable doubt, i.e. that the changes are not due to natural meteorological
178 variability. The meteorological analyses in Supplementary S6 suggest that, while in
179 September 2014 the southern part of the spatial domain shown in Figure 2 is somewhat
180 influenced by anomalous easterlies bringing pollution from the European continent over the
181 easternmost Atlantic Ocean and hence influencing r_{eff} , the perturbations to r_{eff} during October
182 2014 are entirely of volcanic origin.

183 MODIS and HadGEM3 show a similar spatial distribution and magnitude for October for the
184 perturbation in cloud droplet number concentration (ΔN_d), but a smaller ΔN_d in MODIS than
185 in HadGEM3 for September 2014 (Supplementary S7.2). Once r_{eff} is reduced, the
186 autoconversion process whereby cloud droplets grow to sufficient size to form precipitation

187 may be inhibited, leading to clouds with increased liquid water path³. The cloud optical
188 depth, τ_{cloud} , is related to r_{eff} and LWP and the density of water (ρ) by the approximation:

$$189 \quad \tau_{cloud} \cong \frac{3LWP}{2\rho r_{eff}} \quad (1)$$

190 We use HadGEM3 to assess the detectability of perturbations against natural variability. Two
191 different methods are pursued using the nudged model; firstly, assessing model simulations
192 with and without the emissions from the eruption for the year 2014 (HOL₂₀₁₄-NO_HOL₂₀₁₄),
193 and secondly assessing model simulations including emissions from Holuhraun for 2014
194 against simulations for 2002-2013 (HOL₂₀₁₄-NO_HOL₂₀₀₂₋₂₀₁₃). While the former method
195 allows the ‘cleanest’ assessment of the impacts of the eruption (as the meteorology is
196 effectively identical and meteorological variability is removed), the second method allows
197 assessment of the statistical significance against the natural meteorological variability. This
198 provides an assessment that is directly comparable to observations and can be used to
199 effectively isolate signal from noise³⁷ (Supplementary S7).

200

201 *****Insert Figure 4 here*****

202

203 Figure 4 shows that ΔAOD , ΔN_d , and Δr_{eff} are statistically significant at 95% confidence
204 across the majority of latitudes. The fact that the simulations from [HOL₂₀₁₄-NO_HOL₂₀₁₄]
205 and [HOL₂₀₁₄-NO_HOL₂₀₀₂₋₂₀₁₃] are similar for these variables again indicates that the
206 impacts of natural meteorological variability on these variables is small (i.e. NO_HOL₂₀₁₄ \approx
207 NO_HOL₂₀₀₂₋₂₀₁₃). For ΔLWP , no statistically significant changes are evident at either 95% or
208 67% confidence, suggesting that meteorological variability provides a far stronger control on
209 cloud LWP than aerosol (Supplementary S7.3). With ΔLWP being due to meteorological
210 noise, $\Delta \tau_{cloud}$ is driven by Δr_{eff} and Figure 4e suggests that the perturbations to τ_{cloud} north of
211 around 67°N/57°N, which are significant at the 95%/67% confidence level, are due to the
212 2014-15 Holuhraun eruption. Our simulations suggest that Top of Atmosphere changes in

213 short wave radiation ($\Delta T_{\Delta A_{SW}}$) are unlikely to be detectable at 95% or even 67% confidence
214 when compared to natural variability. More details supporting this assertion are given in
215 Supplementary S7.5 which uses satellite observations of the Earth's radiation budget.

216 We have shown that HadGEM3 is capable of representing observations of aerosol-cloud
217 interactions with a reasonable representation of the perturbation to r_{eff} but minimal
218 perturbation to LWP . To demonstrate the practical value of the study, we repeat the
219 simulations with other models. First, we use HadGEM3 but using the older single moment
220 CLASSIC³⁸ aerosol scheme instead of the new two-moment UKCA/GLOMAP-mode
221 scheme³⁹. We also perform calculations with the NCAR Community Atmosphere Model²⁸
222 (CAM5-NCAR) and the atmospheric component of an intermediate version of the Norwegian
223 Earth System Model⁴⁰ (CAM5-Oslo), driven using nominally the same emissions and plume
224 top height. CAM5-NCAR has been used previously in free-running mode to provide an initial
225 estimate of the radiative forcing of the 2014-15 Holuhraun eruption²⁸, but as in the
226 HadGEM3 simulations we run CAM5-NCAR and CAM5-Oslo in nudged mode to simulate
227 the meteorology during the eruption as closely as possible. Figure 5 shows a comparison of
228 Δr_{eff} and ΔLWP derived from HOL₂₀₁₄-NO_HOL₂₀₁₄ simulations from HadGEM3,
229 HadGEM3-CLASSIC, CAM5-NCAR, CAM5-Oslo and MODIS for October. We chose
230 October as the contribution from continental Europe pollution to cloud property anomalies
231 has been shown to be small (Supplementary S4-6-7; Supplementary S8 shows the impacts on
232 cloud properties in September).

233

234 *****Insert Figure 5 here*****

235

236 It is immediately apparent from the first column of Figure 5 that HadGEM3 using UKCA,
237 CAM5-NCAR, and CAM5-Oslo are able to accurately model the impact on Δr_{eff} , while
238 HadGEM3-CLASSIC produces an impact that is too strong when compared to the MODIS
239 observations owing to the single moment nature of the aerosol scheme (Supplementary S9).

240 For ΔLWP , as we have seen from the multi-year analysis of MODIS (Supplementary Fig.
241 S7.3), the meteorological variability is the controlling factor. Even with meteorological
242 variability suppressed in these [HOL₂₀₁₄-NO_HOL₂₀₁₄] results, HadGEM3 using UKCA
243 shows only a very limited increase in LWP (Fig. 5f), HadGEM3-CLASSIC and CAM5-Oslo
244 show a progressively more significant response whereas CAM5-NCAR shows a much larger
245 response (Fig. 5h).

246 It is insightful to examine the influence of the eruption on precipitation in both observations
247 and models using a similar analysis (Supplementary S10). We observe that there is little
248 impact on precipitation indicating that the cloud system readjusts to a new equilibrium with
249 little impact on either LWP or precipitation. The larger response in CAM5-NCAR ($\Delta LWP >$
250 16 g.m^{-2}) is not supported by the MODIS observations where the 2002-2013 domain mean
251 standard deviation in ΔLWP is $\sim 4.5 \text{ g.m}^{-2}$. Thus, we are able to use the eruption to evaluate
252 the models: HadGEM3 using UKCA and CAM5-Oslo perform in a manner consistent with
253 the MODIS observations while HadGEM3-CLASSIC and CAM5-NCAR do not. Moreover,
254 the fact that changes in LWP are not detectable above natural variability suggests that
255 aerosol-cloud interactions beyond the impact on r_{eff} are small (i.e. net second indirect effects
256 are small).

257 The effective radiative forcing (ERF) from the event may be estimated from the difference
258 between the top of atmosphere net irradiances from simulations including and excluding the
259 volcanic emissions. The global ERF from HadGEM3 over the September-October 2014
260 period is estimated at -0.21 W.m^{-2} . Tests using an offline version of the radiation code reveal
261 that the presence of overlying ice-cloud weakens the ERF by approximately 20%
262 (Supplementary S11).

263 We also investigate whether a fissure eruption of this magnitude could have a more
264 significant radiative impact if the timing/location of the eruptions were different
265 (Supplementary S12). Our simulations suggest that for contrasting scenarios the global ERF
266 would *i*) strengthen to -0.29 W.m^{-2} (+40%) if the eruption commenced at the beginning of

267 June, *ii*) strengthen to -0.49 W.m^{-2} (+140%) if the fissure eruption had occurred in an area of
268 South America where it could affect clouds in a stratocumulus-dominated regime, *iii*)
269 strengthen to -0.32 W.m^{-2} (+55%) if the eruption had occurred in pre-industrial times when
270 the background concentrations of aerosols was reduced¹⁸ indicating that climatic impact of
271 fissure eruptions such as Laki⁴¹ in 1783-1784 would not have been as large if it had occurred
272 in the present day.

273 Many studies^{9,11,42,43} suggest that cloud adjustments may be dependent upon meteorological
274 regime, so we ask whether the cloud *LWP* invariance observed near Holuhraun is simply a
275 special case. We have reproduced the cloud regimes analysis derived from satellite
276 measurements presented in a recent study⁴⁴. We find that, when examining the 2014-15
277 eruption at Holuhraun, we are far from examining a meteorological ‘special case’, in fact
278 rather the opposite (Supplementary S13); we are examining a region that contains the whole
279 spectrum of liquid-dominated cloud regimes and deducing that, overall, the impact on *LWP* is
280 minimal.

281 To further support our conclusion, we report results from a different event (Mount Kilauea,
282 Hawaii, Supplementary S14), which degassing rate significantly increased during June-
283 August 2008. The outflow of the plume affected the surrounding trade maritime
284 cumuli^{24,45,46}, increasing the SW reflectance; the causal interpretations of this in the literature
285 have varied^{24,46}. affecting the surrounding trade maritime cumuli^{24,45,46} and increased the SW
286 reflectance in the outflow of the plume, although with different causal interpretations^{24,46}.
287 Again, *LWP* does not vary, either in the AMSR-E data⁴⁶ or in the MODIS monthly retrievals
288 (Supplementary S14) which again suggests *LWP* insensitivity in the trade cumulus regime as
289 well. Thus, for a very different meteorological environment dominated by very different
290 cloud regimes, similar conclusions emerge.

291

292 **4. Discussion and Conclusion** (507 words of referenced text):

293 The 2014-15 eruption at Holuhraun presents a unique opportunity to investigate continental-
294 scale aerosol-cloud climatic effects. Using synergistic observations and models driven by an
295 empirical estimate of SO₂ emissions³³ we simulate spatial distributions of SO₂ that compare
296 favourably with satellite observations. The HadGEM3 model is able to predict an impact
297 from aerosol-cloud interactions of similar magnitude to the signal found in the MODIS data.
298 Our analysis further highlights that cloud properties are largely unaffected by the eruption
299 beyond the impact on r_{eff} .

300 We repeated the experiment with two additional GCMs and show that HadGEM3 using
301 UKCA, CAM5-NCAR and CAM5-Oslo are able to capture the magnitude of the observed
302 impacts on r_{eff} despite the lack of explicit representation of processes such as sub-cloud
303 updraft velocities and entrainment, enhancing our confidence in GCMs' ability in predicting
304 the aerosol first indirect effect. However, in line with recent work¹⁶, modelled responses in
305 the LWP differ significantly. The fact that cloud adjustments via LWP are not identified in the
306 observations of the 2014-15 eruption at Holuhraun indicates that clouds are buffered against
307 LWP changes^{9-10,12}, providing evidence that models with a low LWP response display a more
308 convincing behaviour. These findings have wide scientific relevance in the field of climate
309 modelling as, in terms of climate forcing, they suggest that aerosol second indirect effects
310 appear small and climate models with a significant LWP feedback need reassessment^{15-16,47}.

311 Despite such massive emissions and large anomalies in r_{eff} , we estimate a moderate global-
312 mean radiative forcing of $-0.21 \pm 0.08 \text{ W.m}^{-2}$ (1 standard deviation, Supplementary S15) for
313 September-October which equates to a global annual mean effective radiative forcing of
314 $-0.035 \pm 0.013 \text{ W.m}^{-2}$ (1 standard deviation) assuming that a forcing only occurs in
315 September and October 2014. Global emissions of anthropogenic SO₂ currently total around
316 100 TgSO₂/year and the Intergovernmental Panel on Climate Change^{17,47} suggests a best
317 estimate for the aerosol forcing of -0.9 W.m^{-2} , yielding a forcing efficiency of -0.009
318 $\text{W.m}^{-2}/\text{TgSO}_2$. The emissions for September and October 2014 total approximately 4 TgSO₂,
319 thus the global annual mean radiative forcing efficiency for the 2014-15 eruption at

320 Holuhraun yields a forcing efficiency of $-0.0088 \pm 0.0024 \text{ W.m}^{-2}/\text{TgSO}_2$ (1 standard
321 deviation). The similarity is remarkable, but may be by chance given the modelled sensitivity
322 to emission location and time (Supplementary S12).

323 Our study is not without caveats given that the observations themselves are uncertain owing
324 to the limitations of satellite retrievals. The modelling is not completely constrained owing to
325 the lack of detailed in-situ observations of e.g. the background aerosol concentrations and
326 plume height. We cannot rule out that models showing small *LWP* sensitivity to aerosol
327 emission behave as they do because they lack the resolution to represent fine-scale dynamical
328 feedbacks^{9,12}. Further high-resolution modelling of the 2014-15 Holuhraun eruption is
329 necessary to evaluate more thoroughly how processes such as autoconversion or droplet
330 evaporation plays a role in buffering the aerosol effect^{9,12,48,49}. Bringing many of the different
331 global models together and inter-comparing results of Holuhraun simulations is merited to
332 provide a traceable route for reducing the uncertainty in future climate projections.

333

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459

460 **List of Supplementary Materials:**

461 SUPPLEMENTARY_INFORMATION.docx

462 SI-Cloud-Animation.mp4

463 SI-SO2_animation.mp4

464

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490

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495 NC and DL (MODIS cloud regimes), DPG (estimate of CDNC from MODIS data), TT and MEH
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499

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502 **Figure legends:**

503 **Figure 1. The column loading of sulphur dioxide.** *First column: processed data from HadGEM3*
504 *masked using positive detections of SO₂ from IASI and spatially and temporally coherent plume data*
505 *from HadGEM3. Second column: processed data from IASI re-gridded onto the regular HadGEM3*
506 *grid. The column loading are expressed in Dobson Units (DU), with 1 DU equivalent to*
507 *approximately 0.0285 g[SO₂].m⁻². In each case ‘avg’ represents the average concentration derived*
508 *within the plume.*

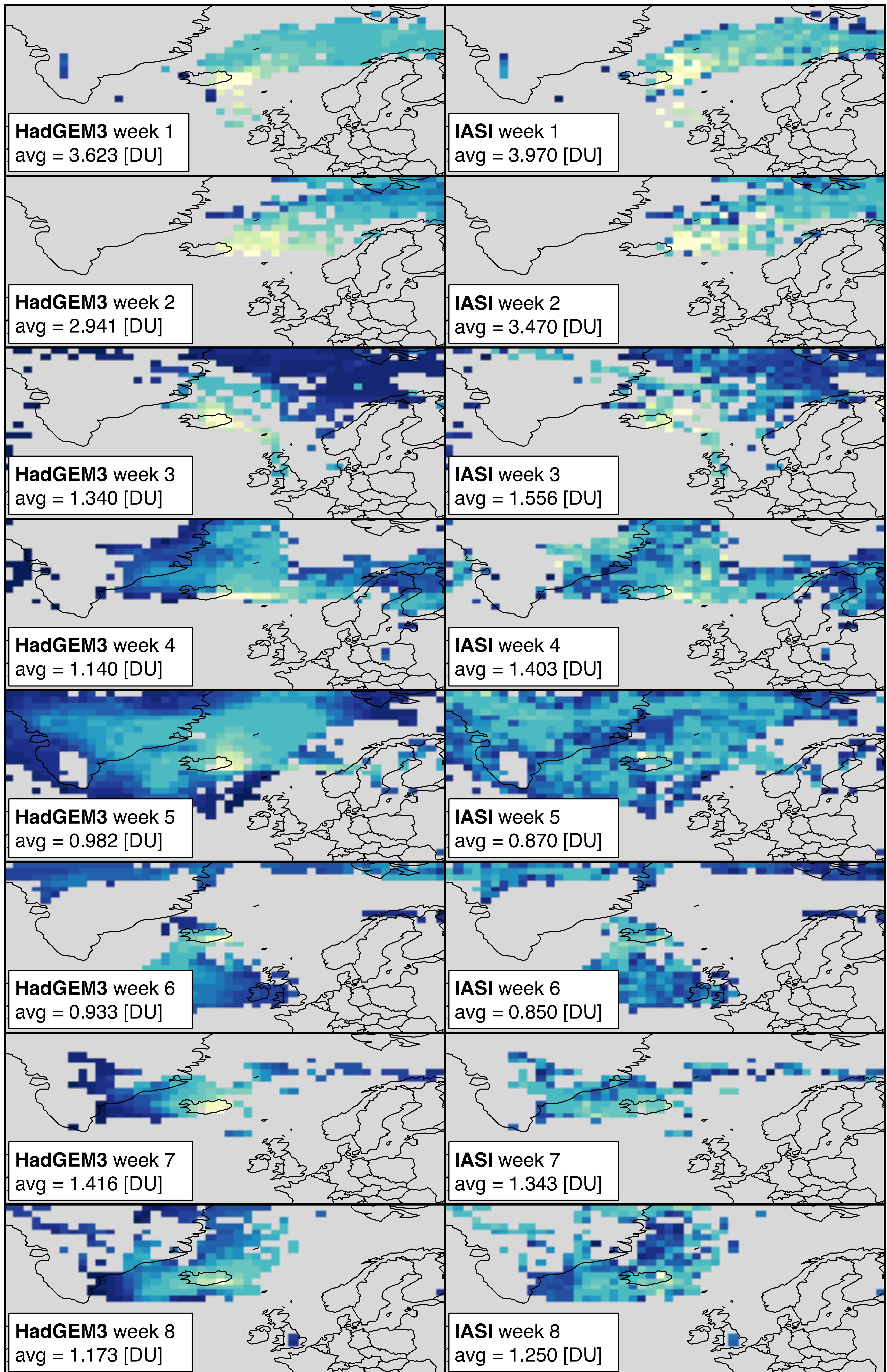
509 **Figure 2. Changes in cloud properties detected by MODIS AQUA for October 2014.** *The mean*
510 *changes in (a) cloud droplet effective radius (μm) and (c) liquid water path (g.m⁻²) with*
511 *corresponding zonal means. The probability distributions of absolute cloud droplet effective radius*
512 *(b) and liquid water path (d) for the year 2014 (blue) and the 2002-2013 mean (green). Changes*
513 *correspond to the deviation from the 2002-2013 mean. Stippling in a) and c) represent areas of 95%*
514 *confidence level significant perturbation based on a two-tailed Student’s t-test. Grey shading in the*
515 *zonal means represent the standard deviation over 2002-2013.*

516 **Figure 3. Changes in cloud properties modelled by HadGEM3 for October 2014.** *The mean*
517 *changes in (a) cloud droplet effective radius (μm) and (c) liquid water path (g.m⁻²) with*
518 *corresponding zonal means. The probability distributions of absolute cloud droplet effective radius*
519 *(b) and liquid water path (d) for 2014 including (blue) or excluding (gold) the Holuhraun emissions,*
520 *and the 2002-2013 mean (green). Changes correspond to the deviation from the 2002-2013 mean.*
521 *Stippling in a) and c) represent areas of 95% confidence level significant perturbation based on a*

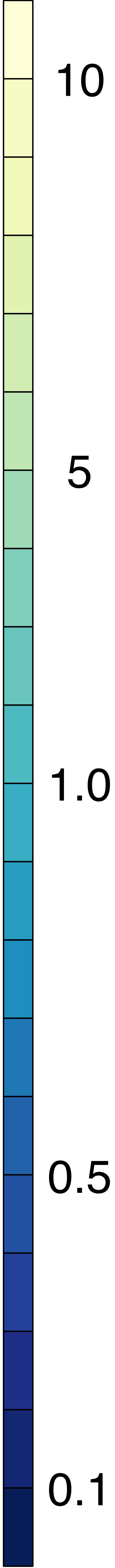
522 two-tailed Student's *t*-test. Grey shading in the zonal means represent the standard deviation over
523 2002-2013.

524 **Figure 4. Modelled perturbations from HadGEM3 using UKCA during the Sept-Oct 2014 period.**
525 Showing perturbations for a) AOD, b) N_d , c) τ_{eff} , d) LWP, e) τ_{cloud} , and f) Top of Atmosphere (ToA) net
526 SW radiation. Zonal means are shown for the 44°N-80°N, 60°W-30°E analysis region. The shaded
527 regions represent the natural variability in the simulations from 2002-2013. Values outside of the
528 light grey (respectively dark grey, bottom row) shaded regions represent significant perturbations at
529 the 95% (respectively 67%) confidence level based on a two-tailed Student's *t*-test. Red lines
530 represent HOL_{2014} minus NO_HOL_{2014} and blue lines represent HOL_{2014} minus $NO_HOL_{2002-2013}$.

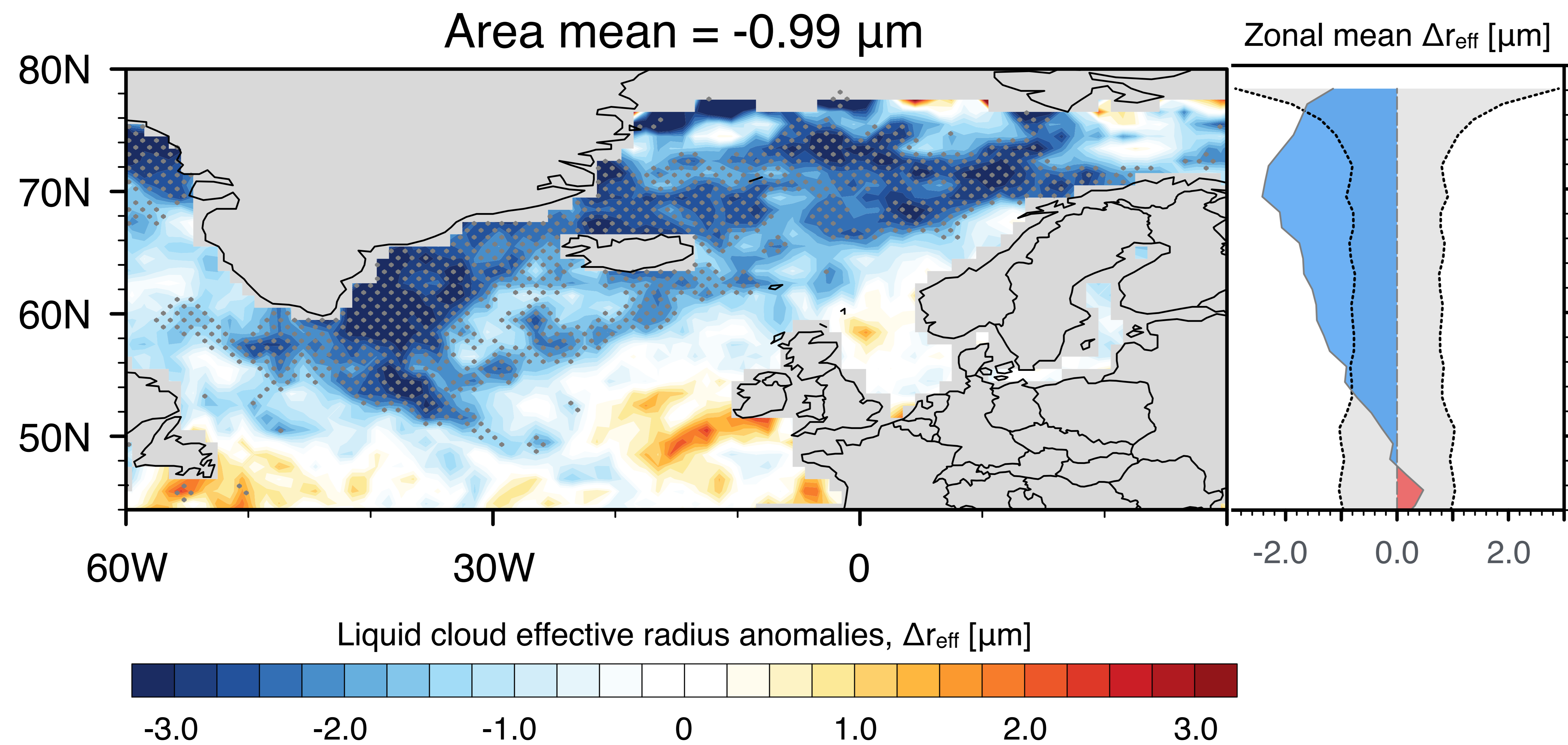
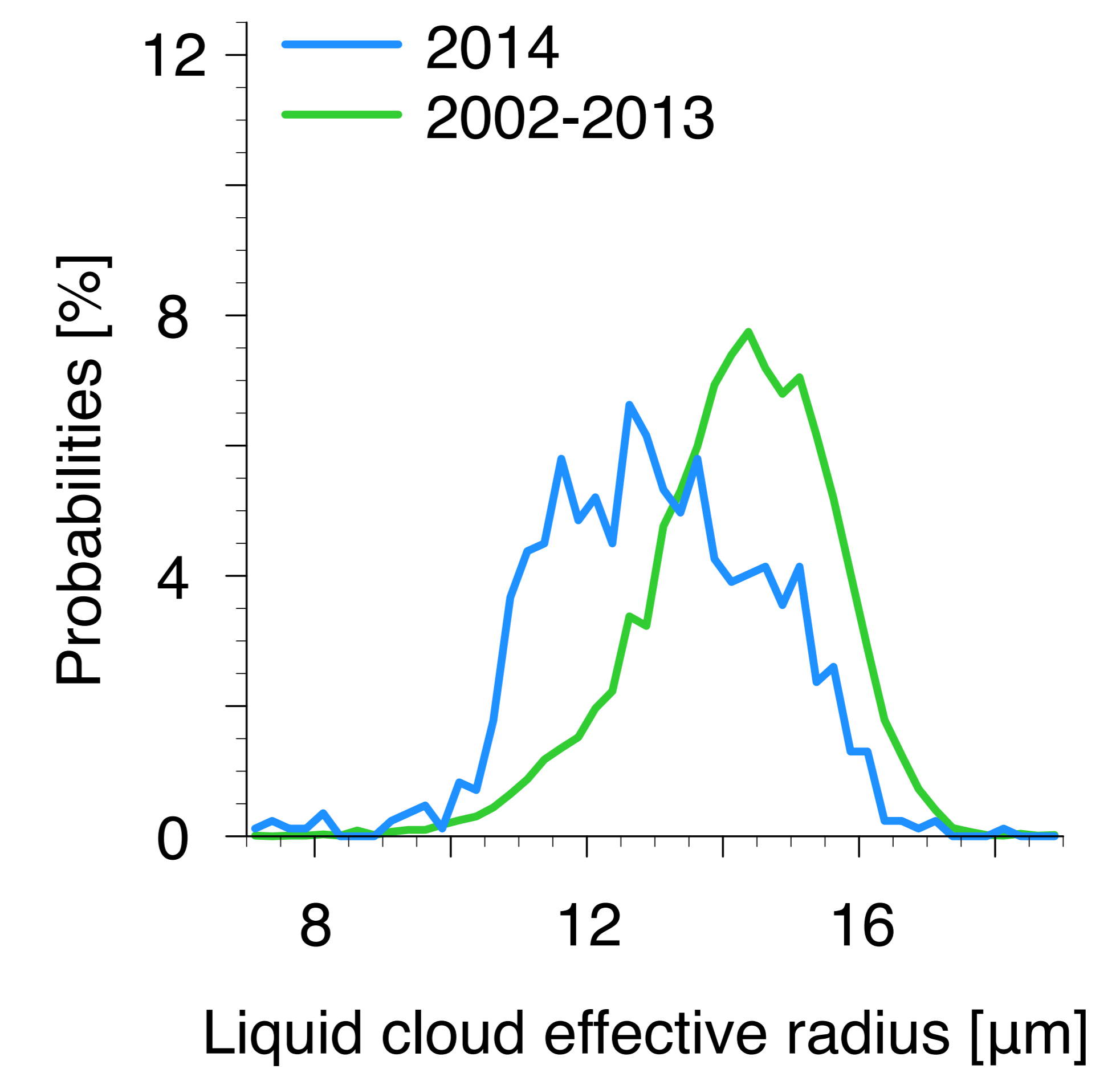
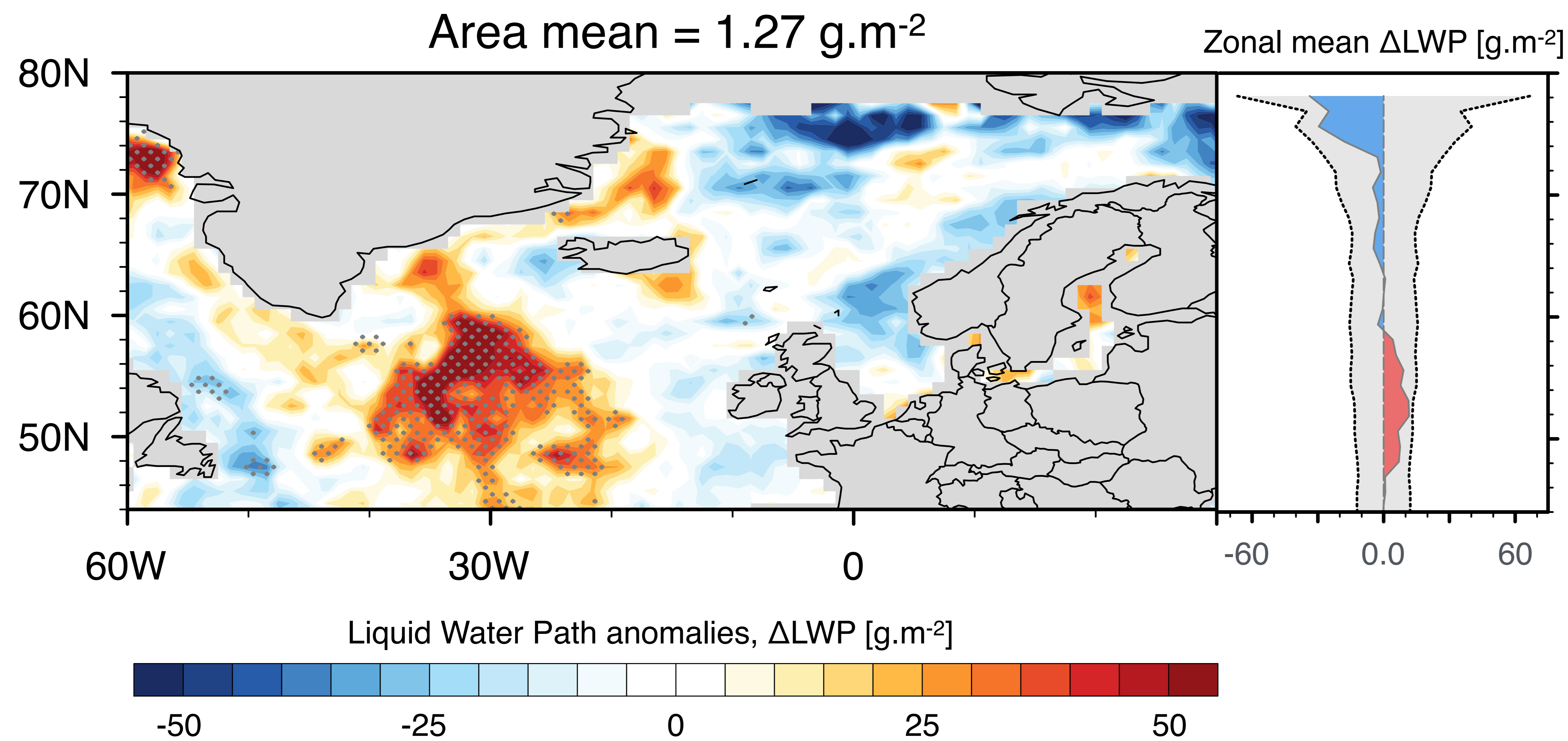
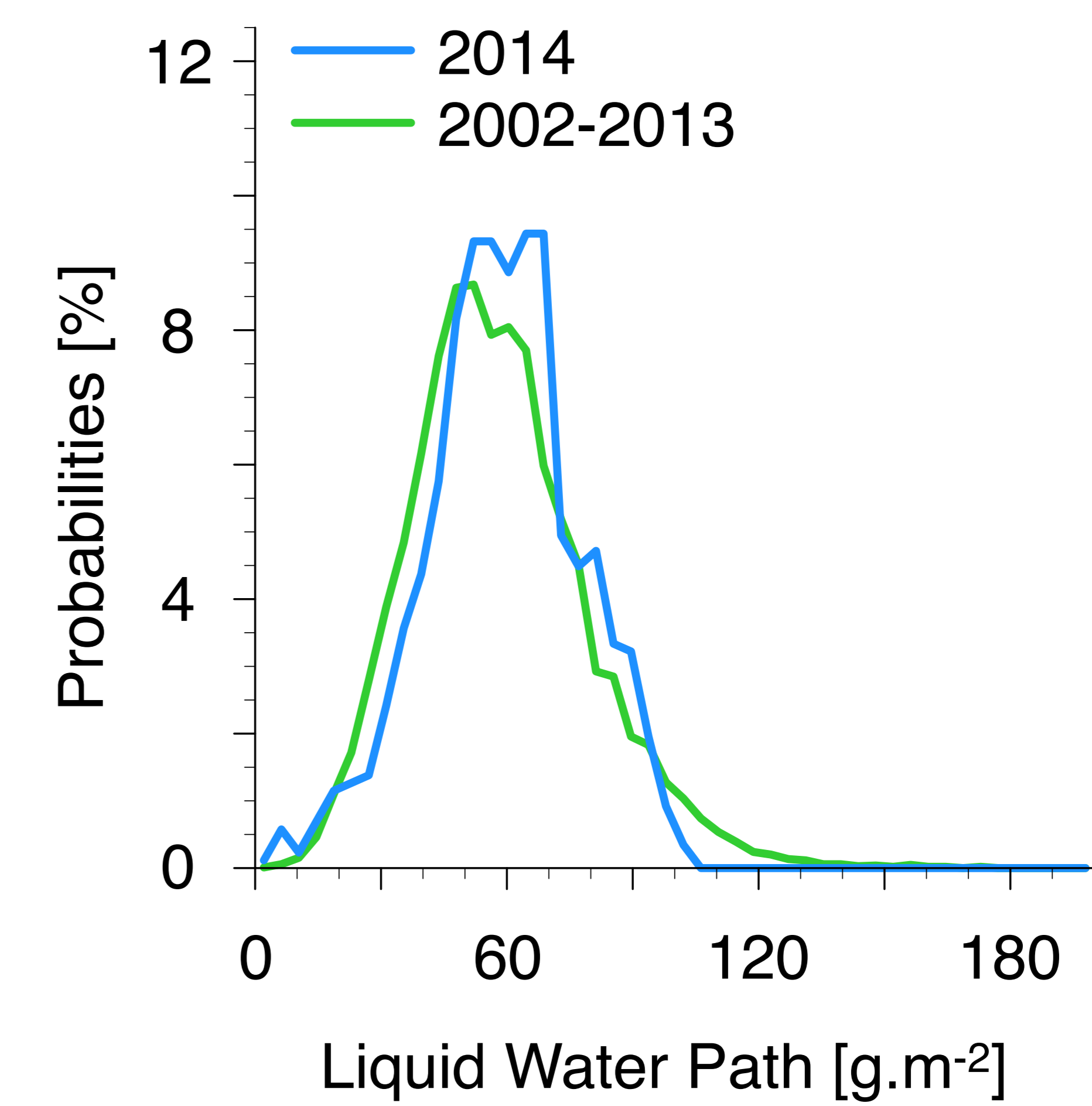
531 **Figure 5. Multi-model estimates of the changes in cloud properties for October 2014.** Left column
532 shows Δr_{eff} (μm) and right column ΔLWP ($\text{g}\cdot\text{m}^{-2}$) determined from HadGEM3 using the 2-moment
533 UKCA/GLOMAP-mode aerosol scheme (first row), HadGEM3 using the single moment CLASSIC
534 aerosol scheme (second row) CAM5-NCAR (third row), CAM5-Oslo (fourth row) and AQUA MODIS
535 (last row). Note that MODIS anomalies show the aerosol impacts plus the meteorological variability
536 while the model simulations show the impact of aerosols only (Supplementary S7).



SO₂ columnar density [DU]

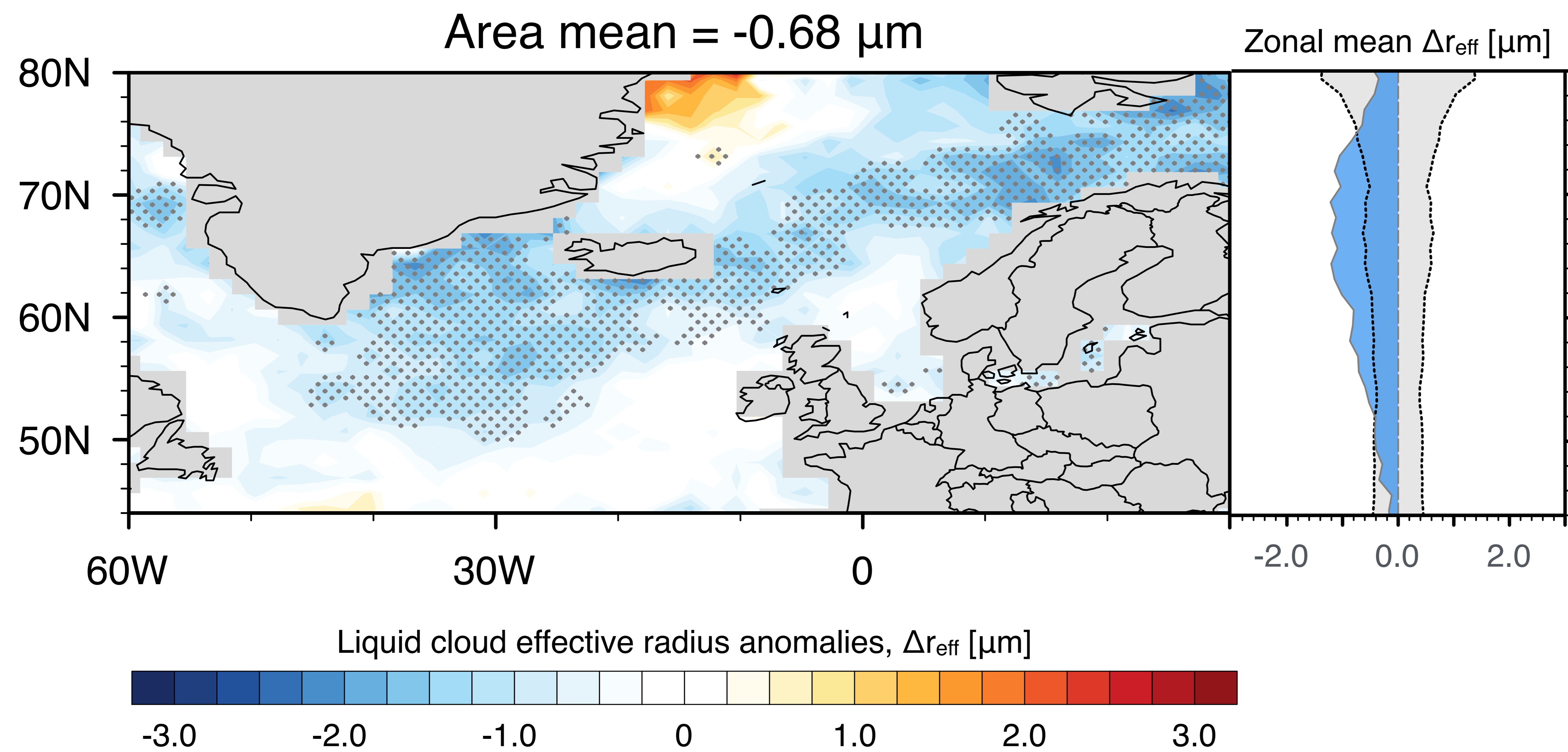


AQUA MODIS - October 2014

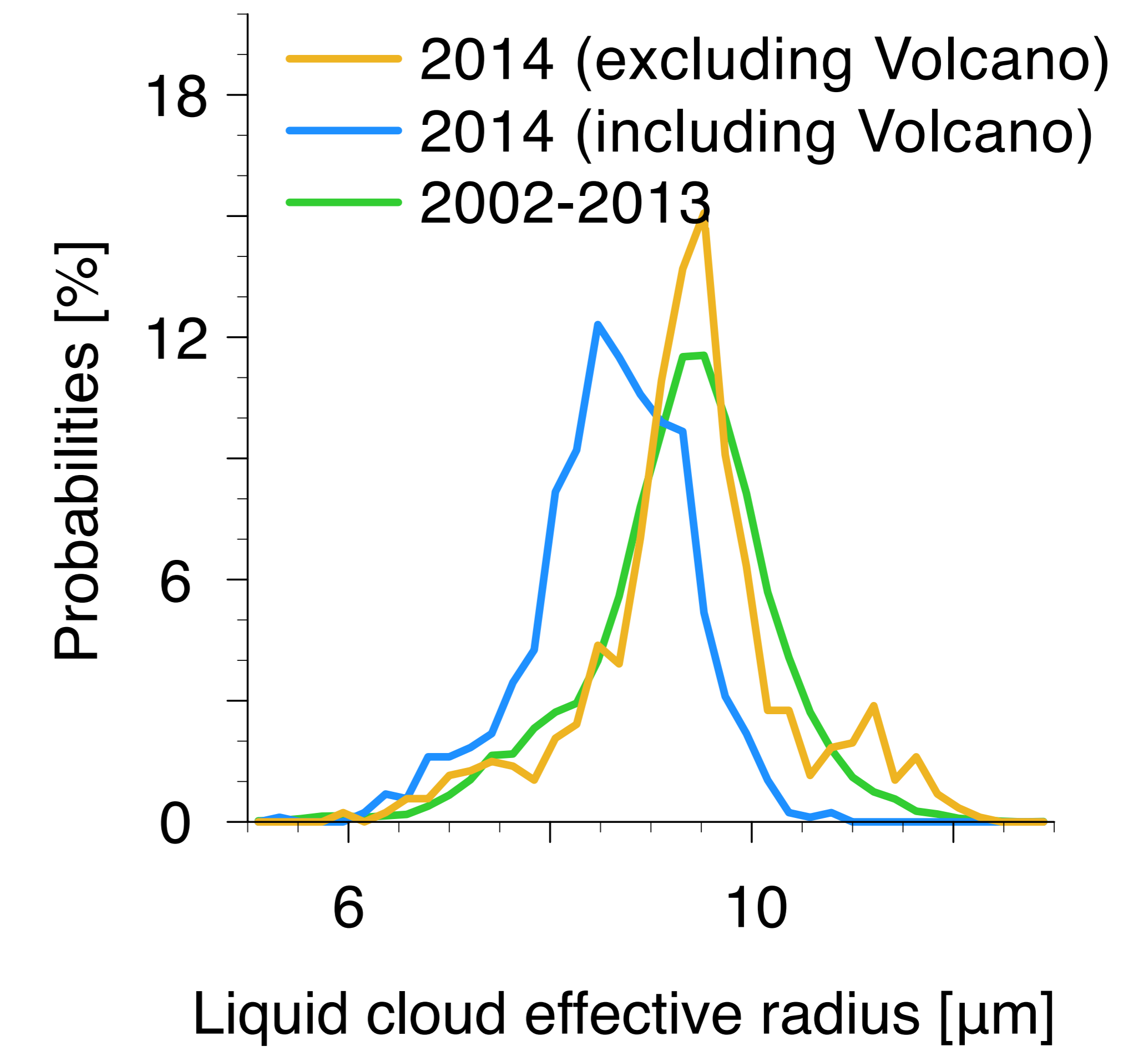
a**b****c****d**

HadGEM3-UKCA - October 2014

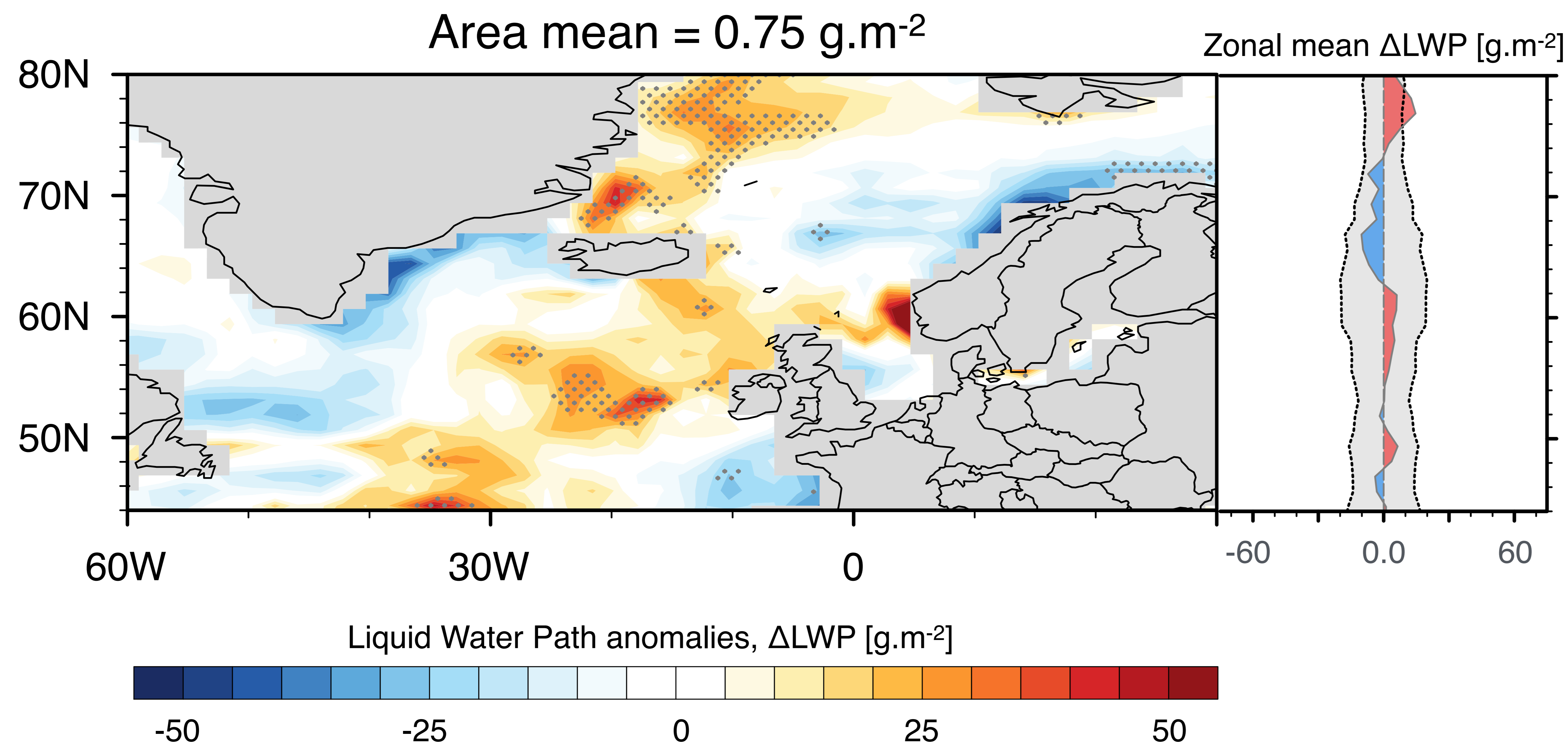
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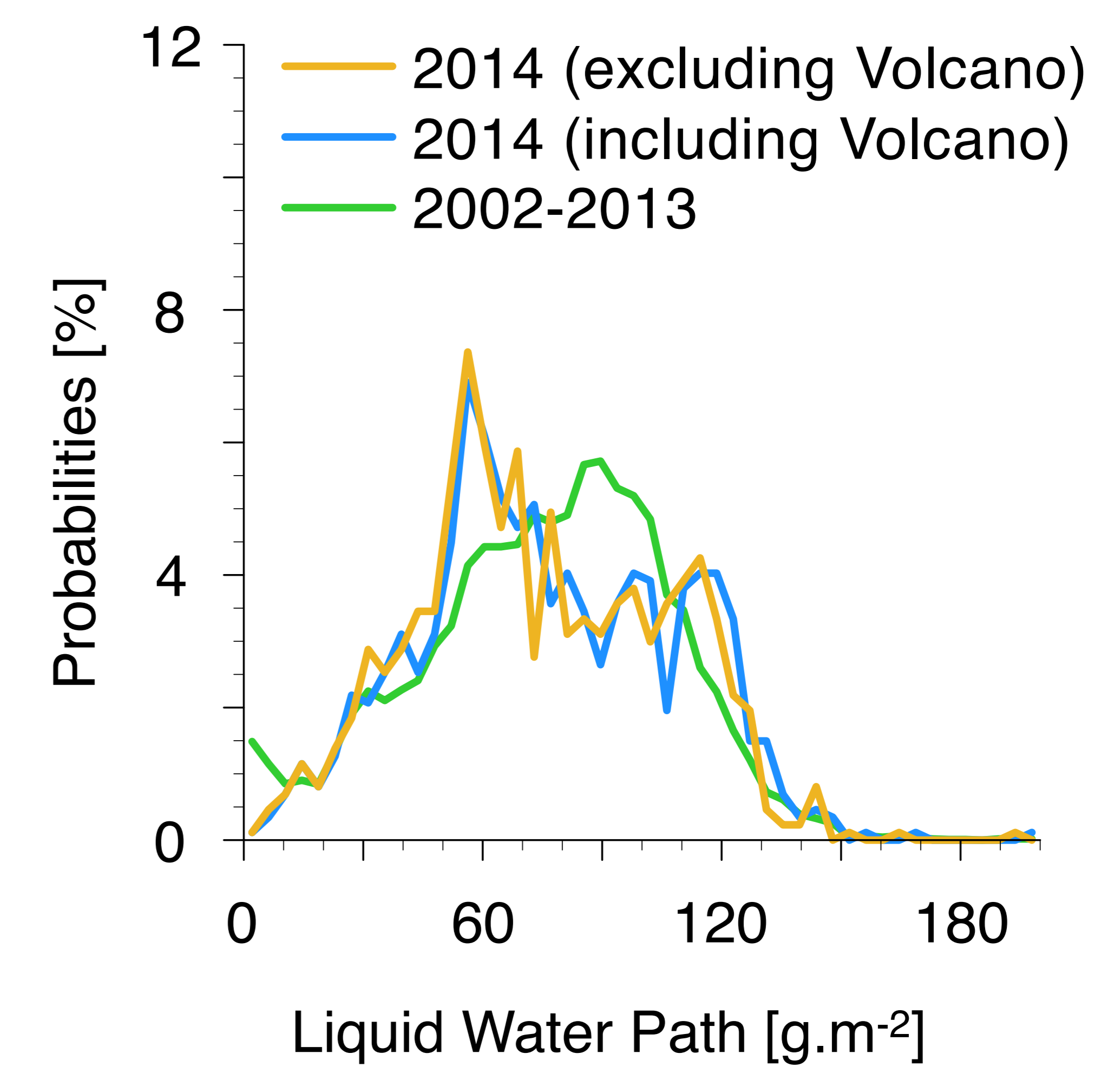
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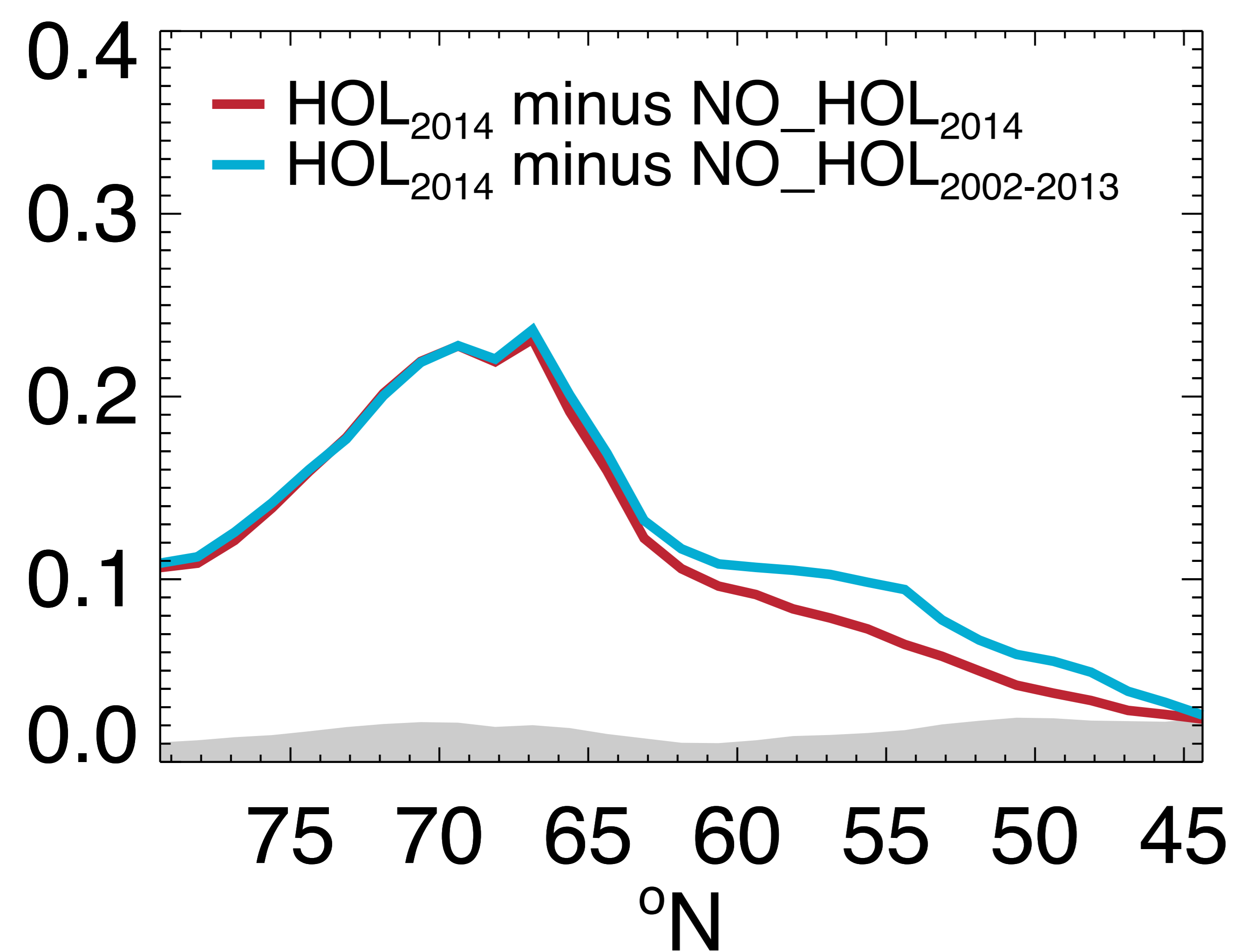
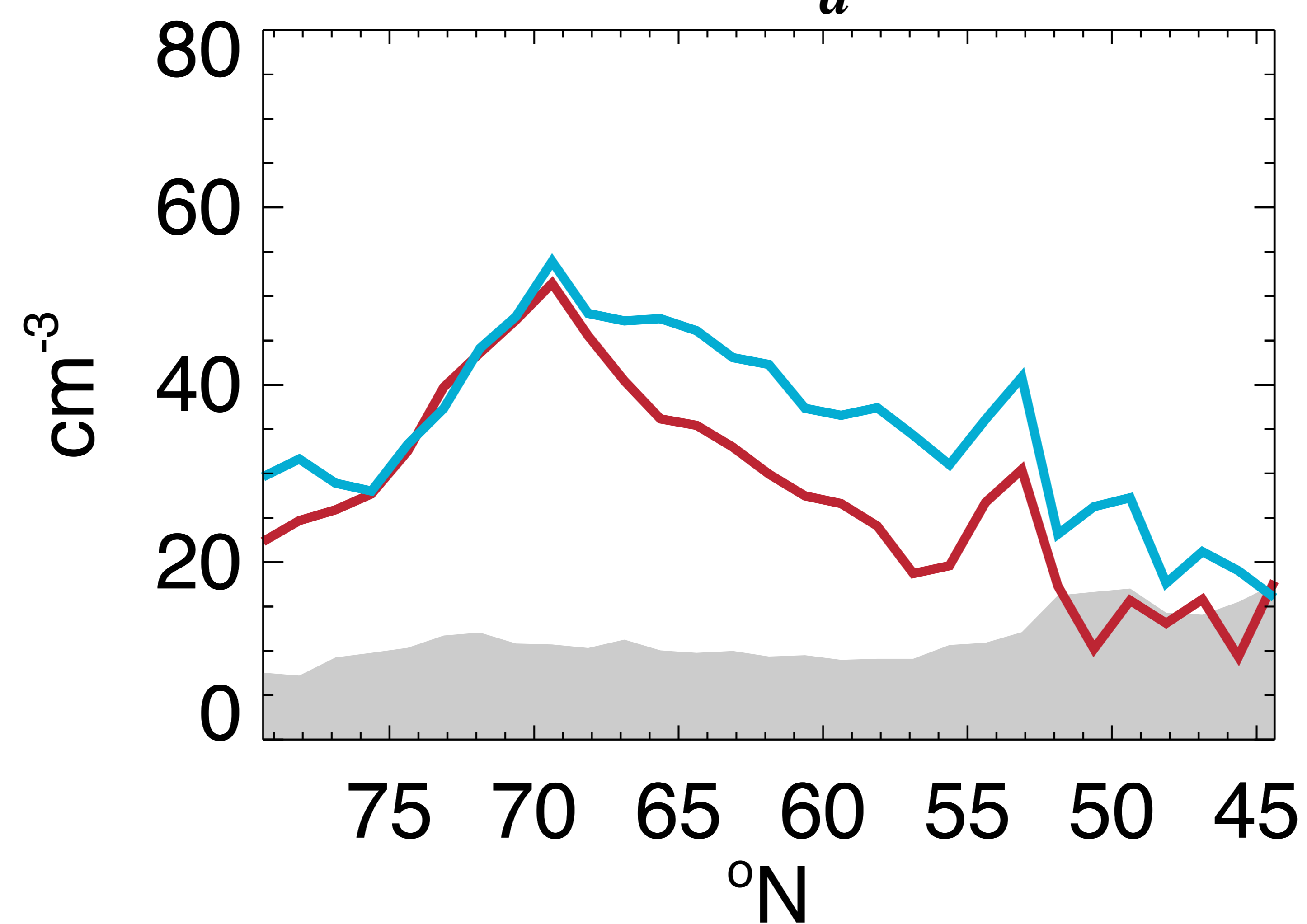
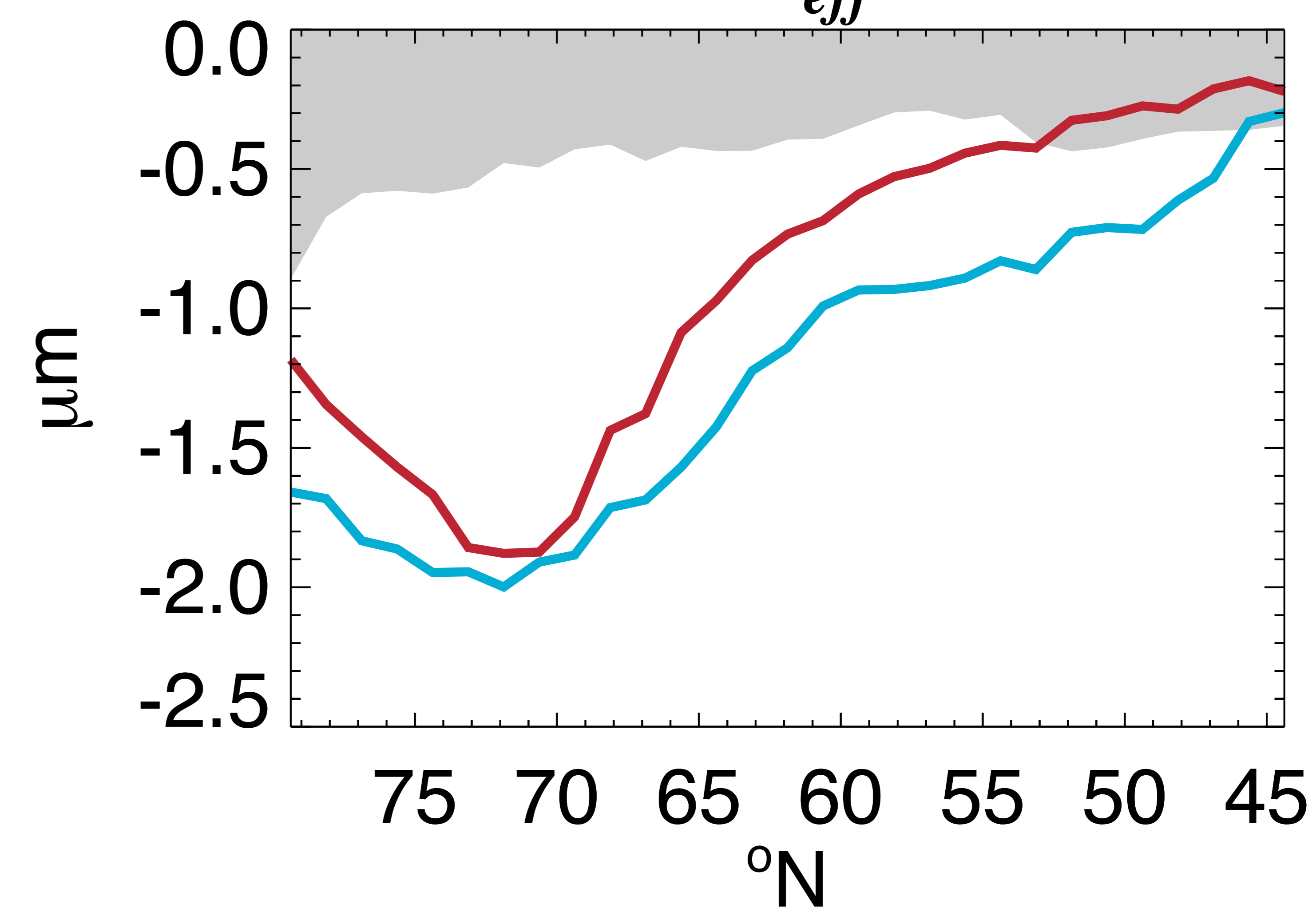
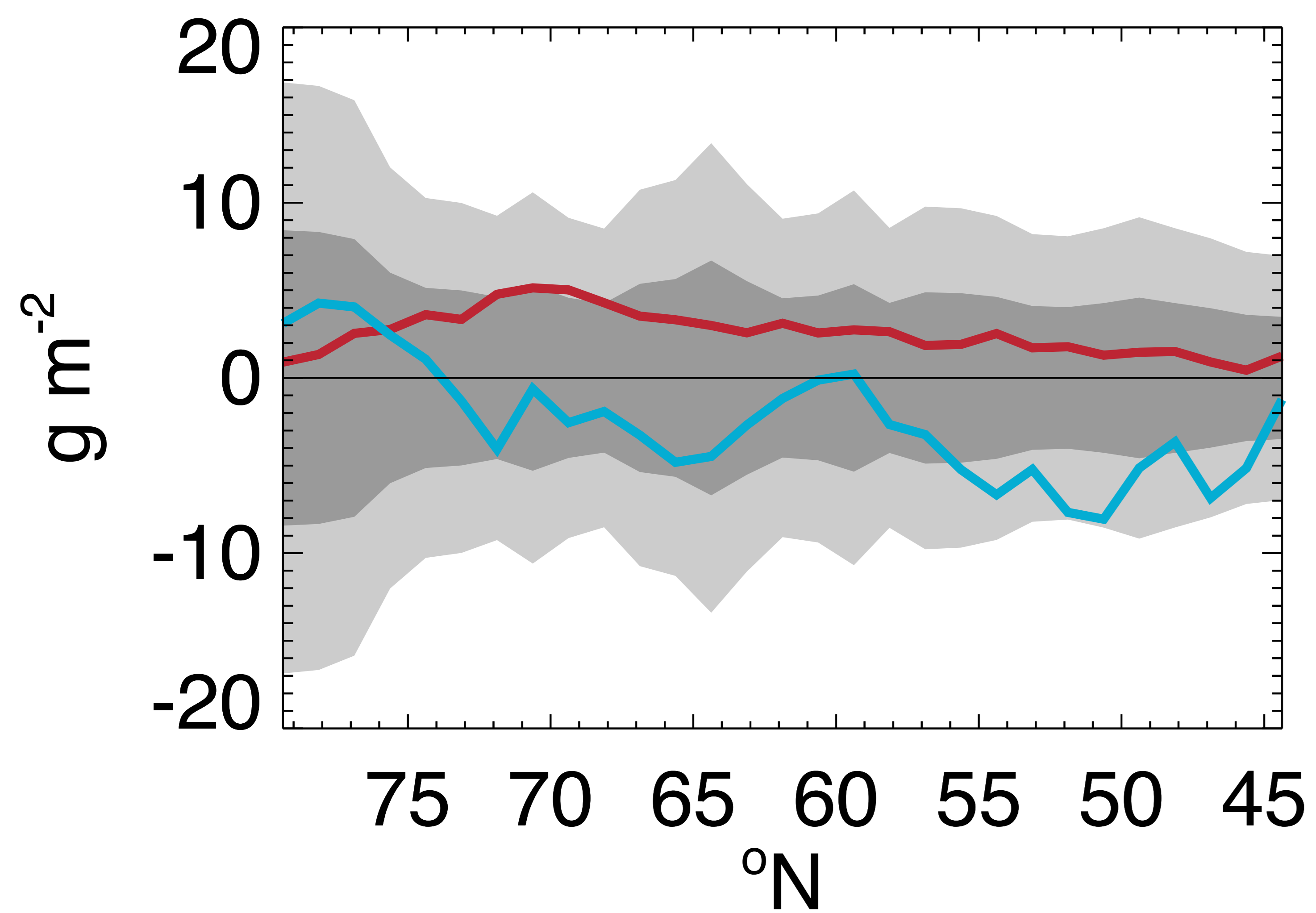
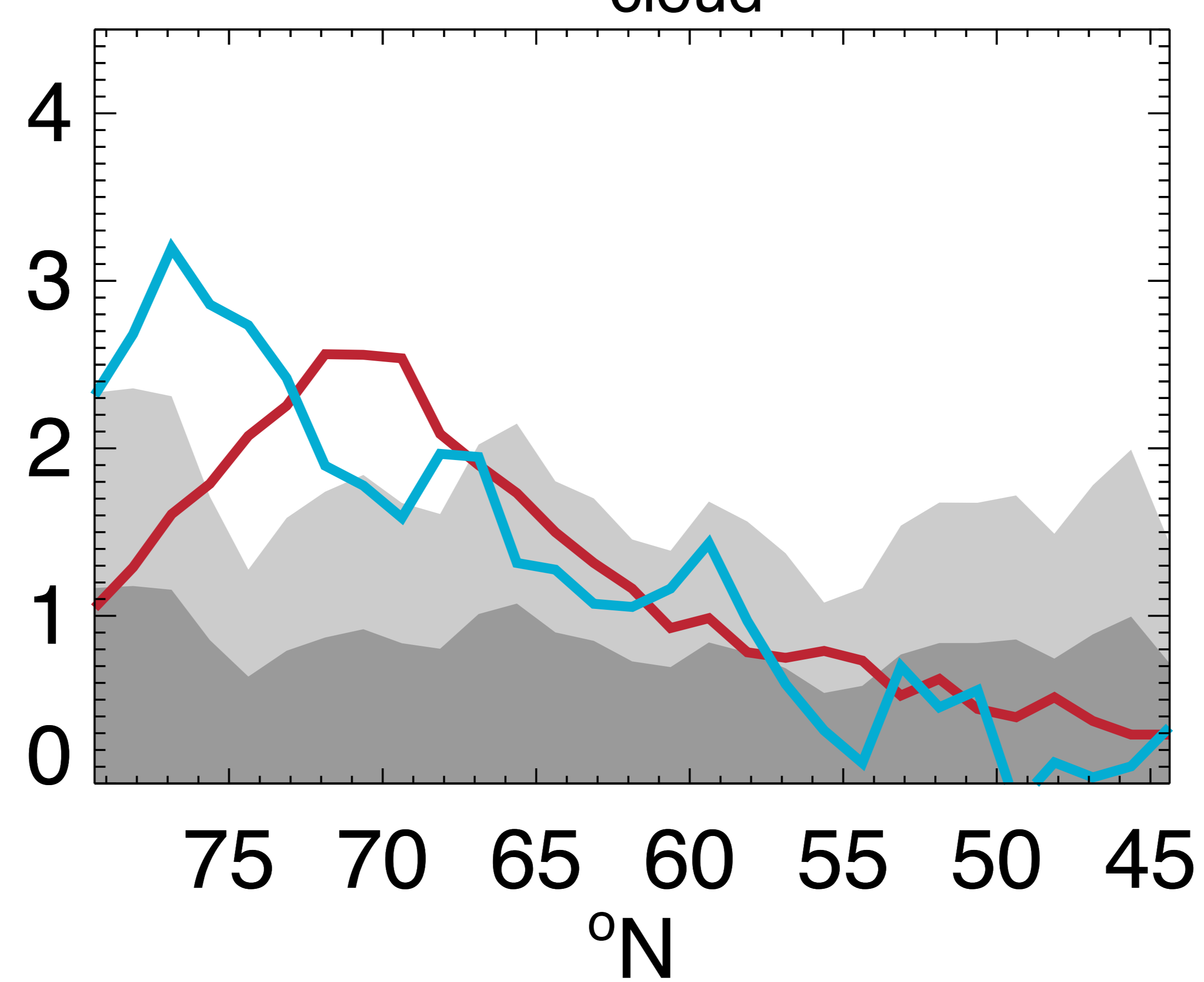
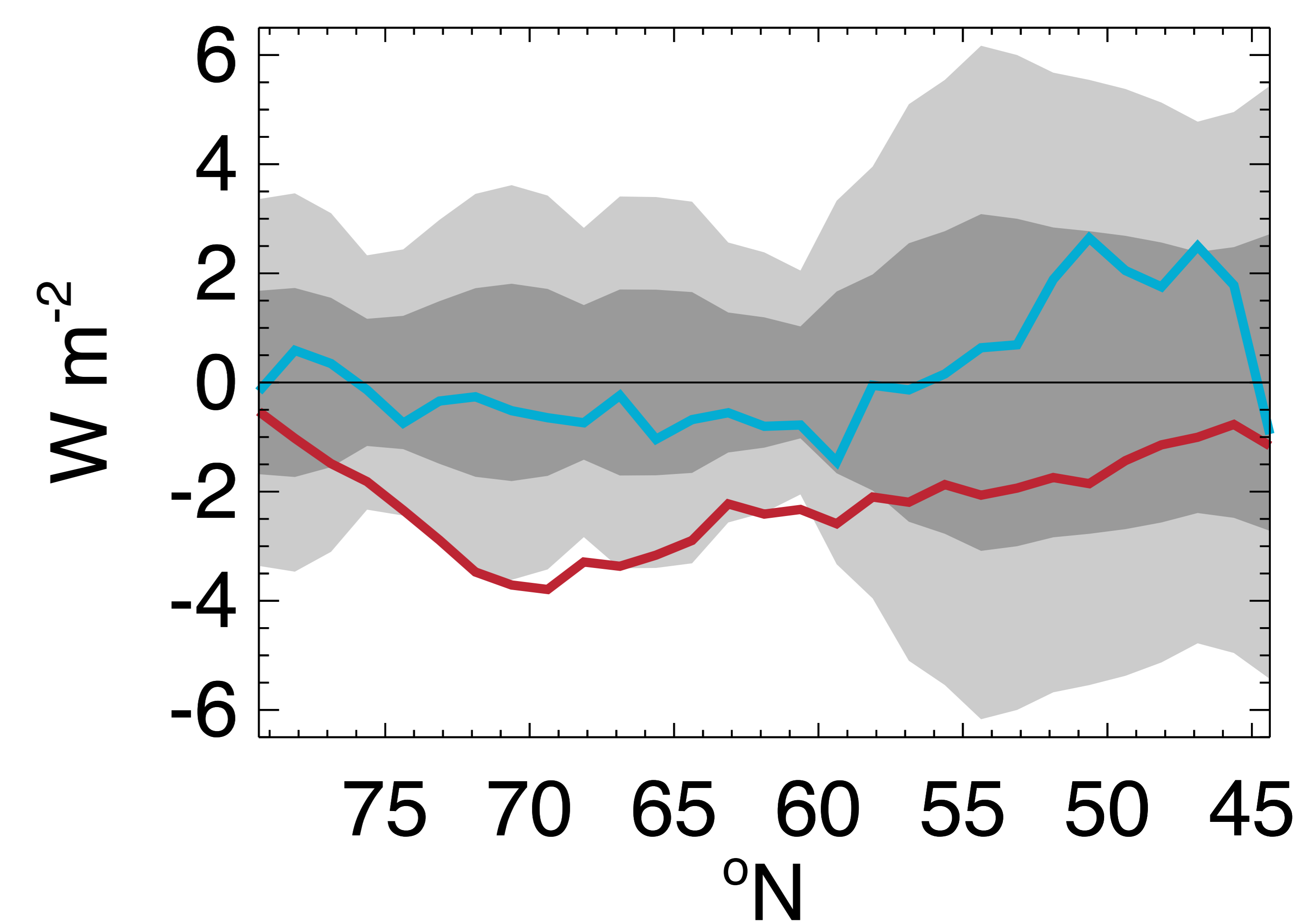


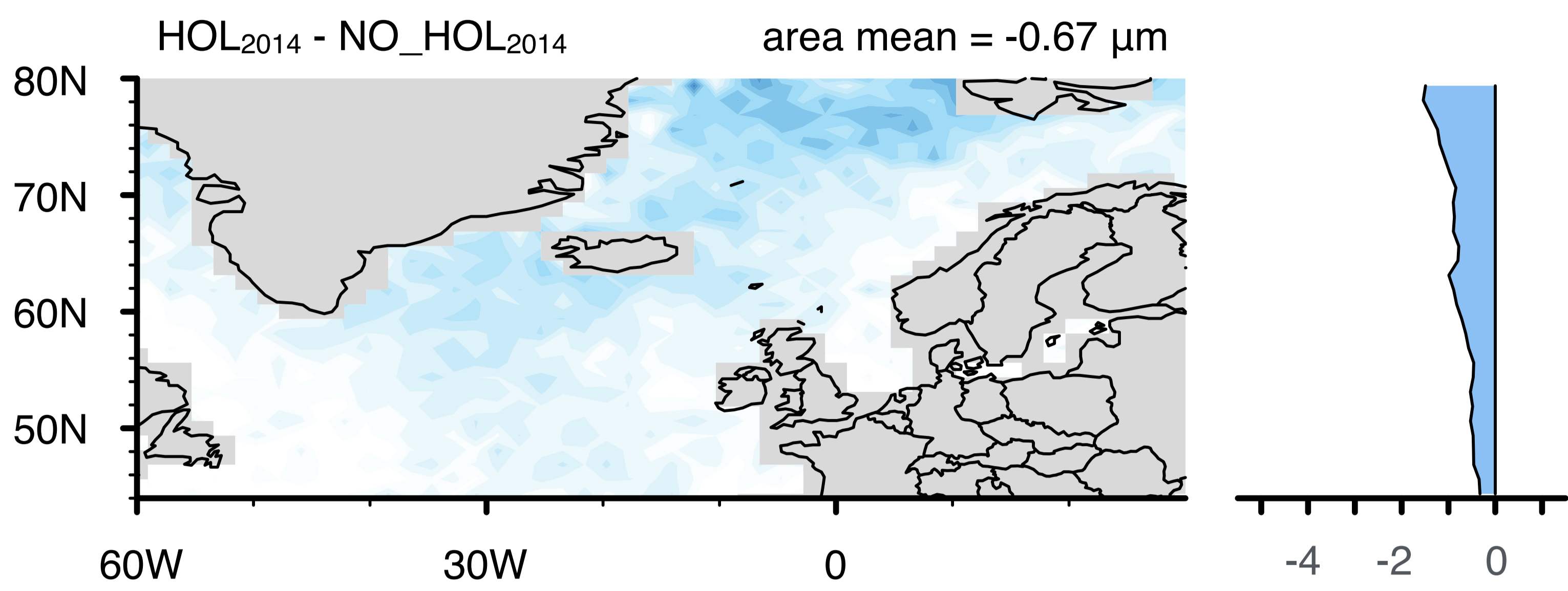
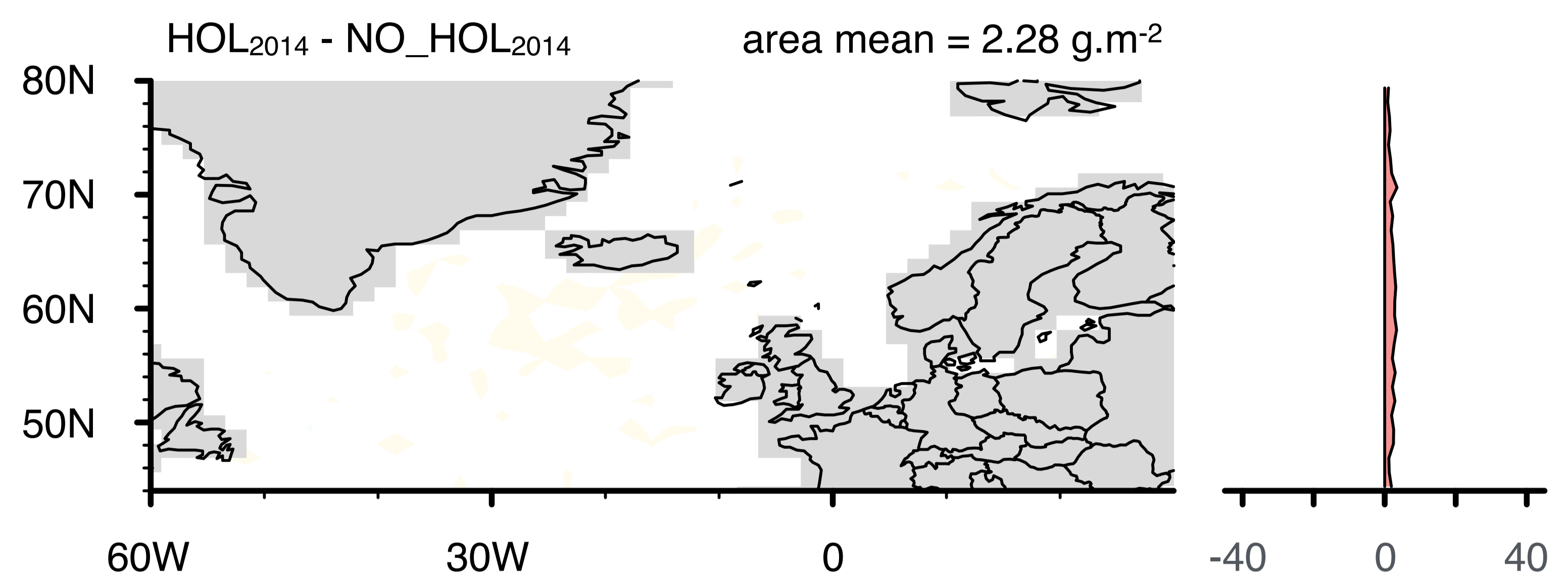
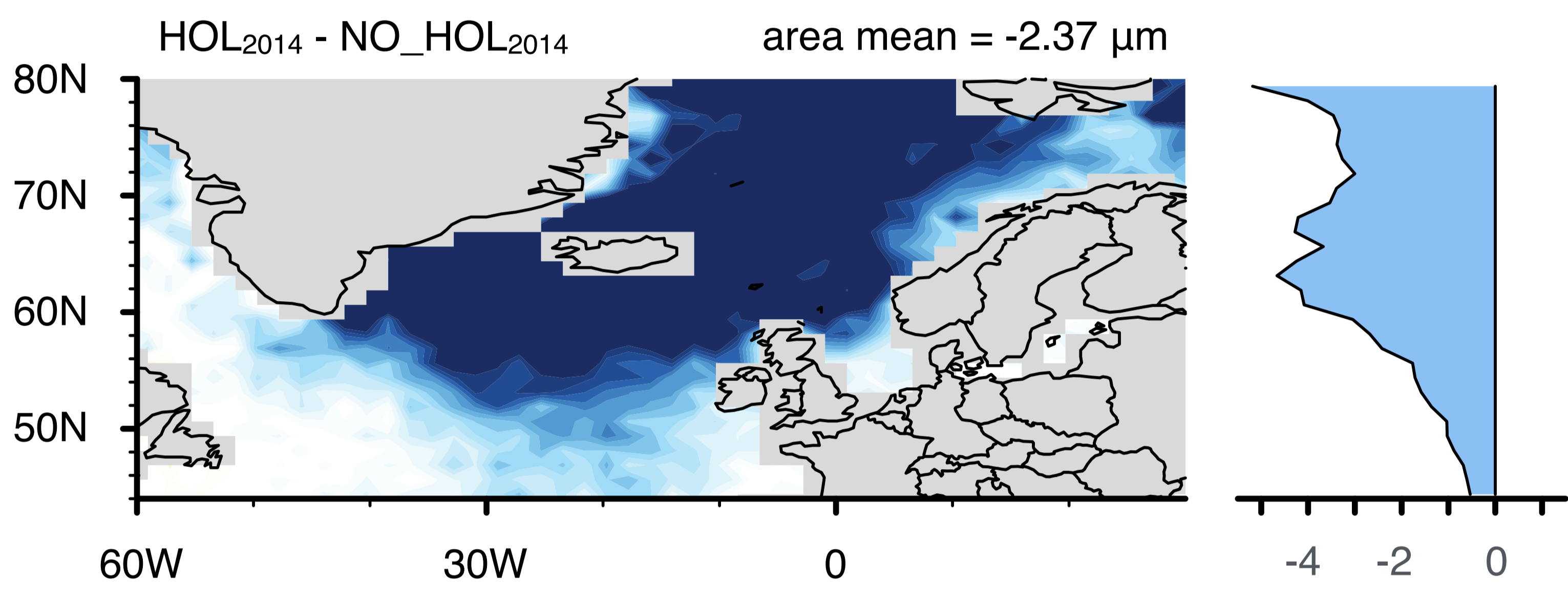
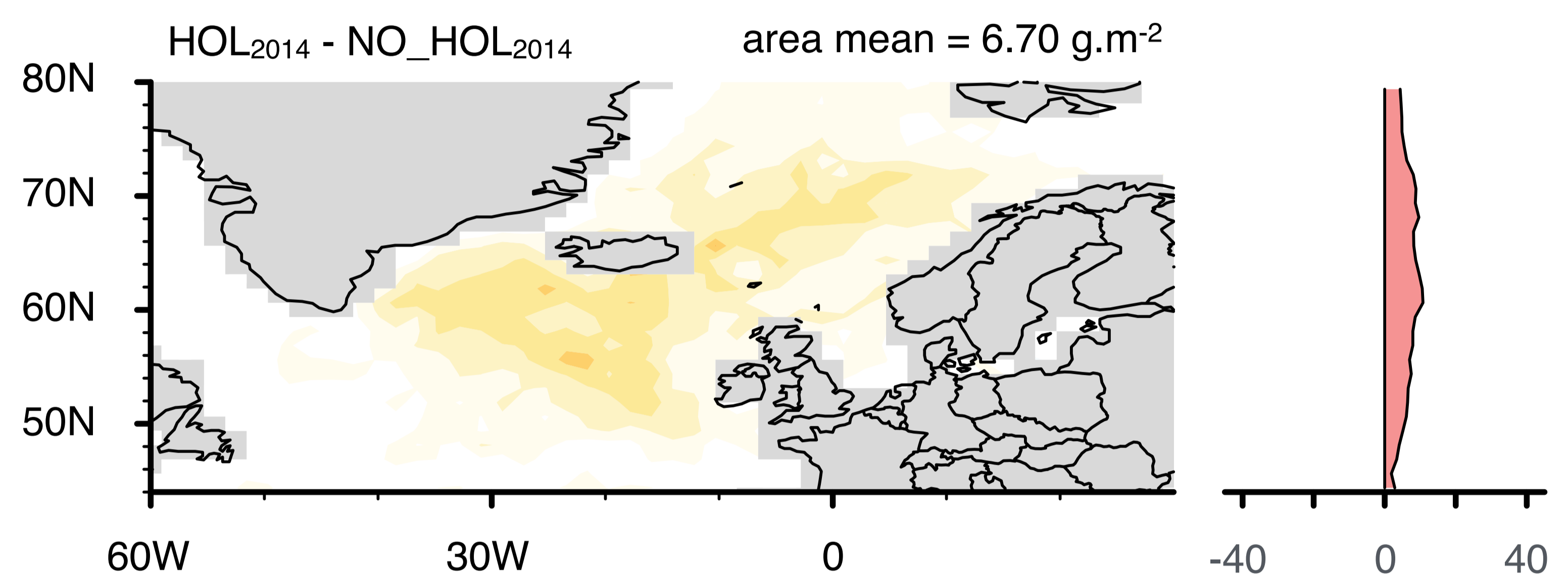
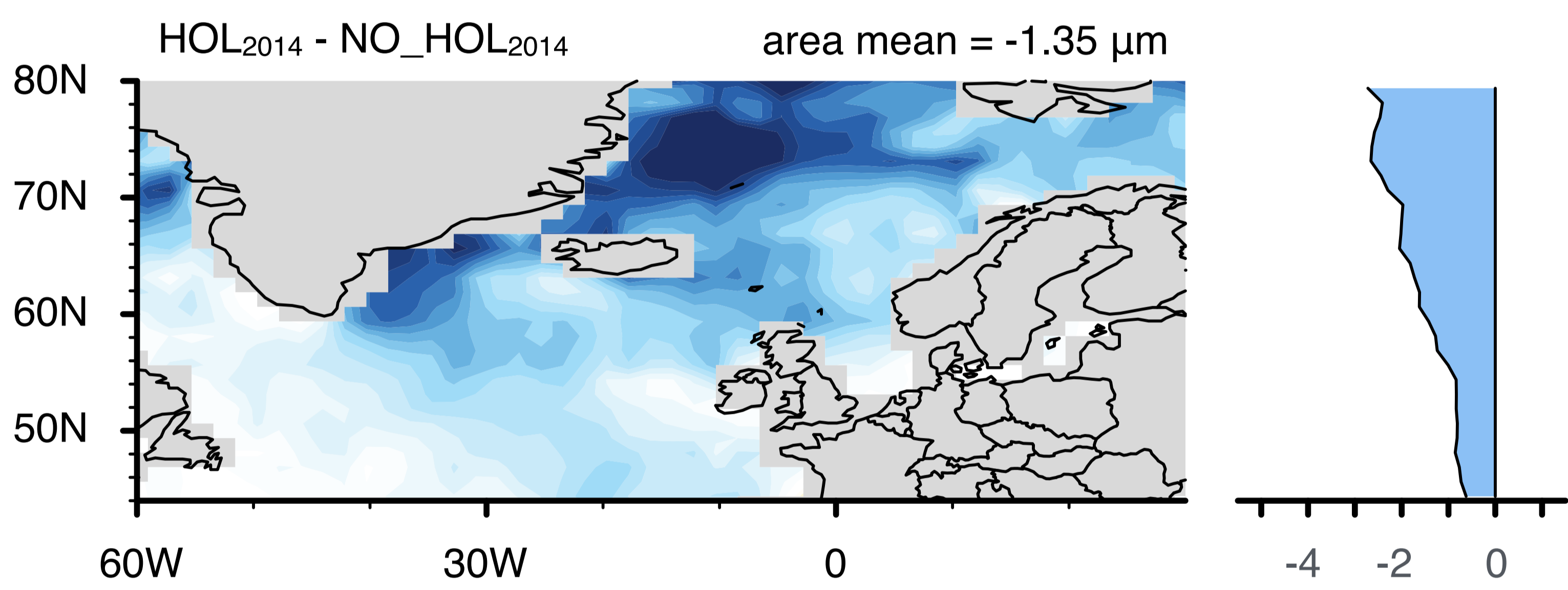
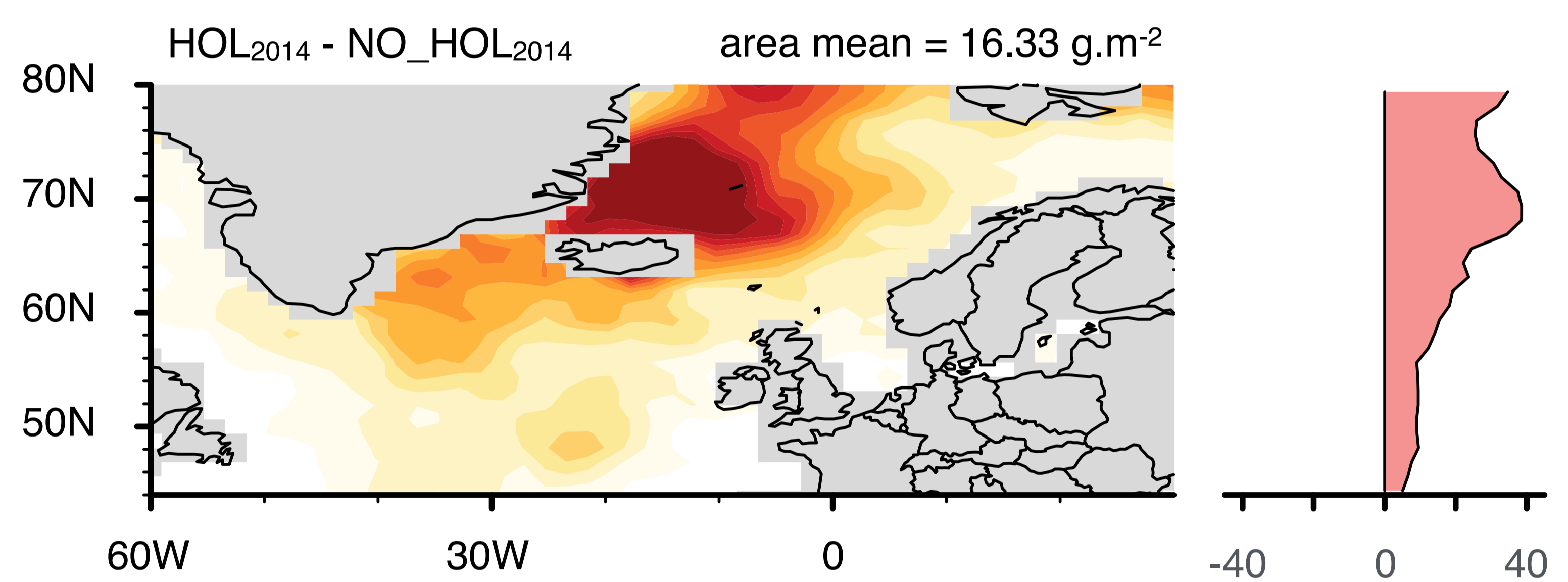
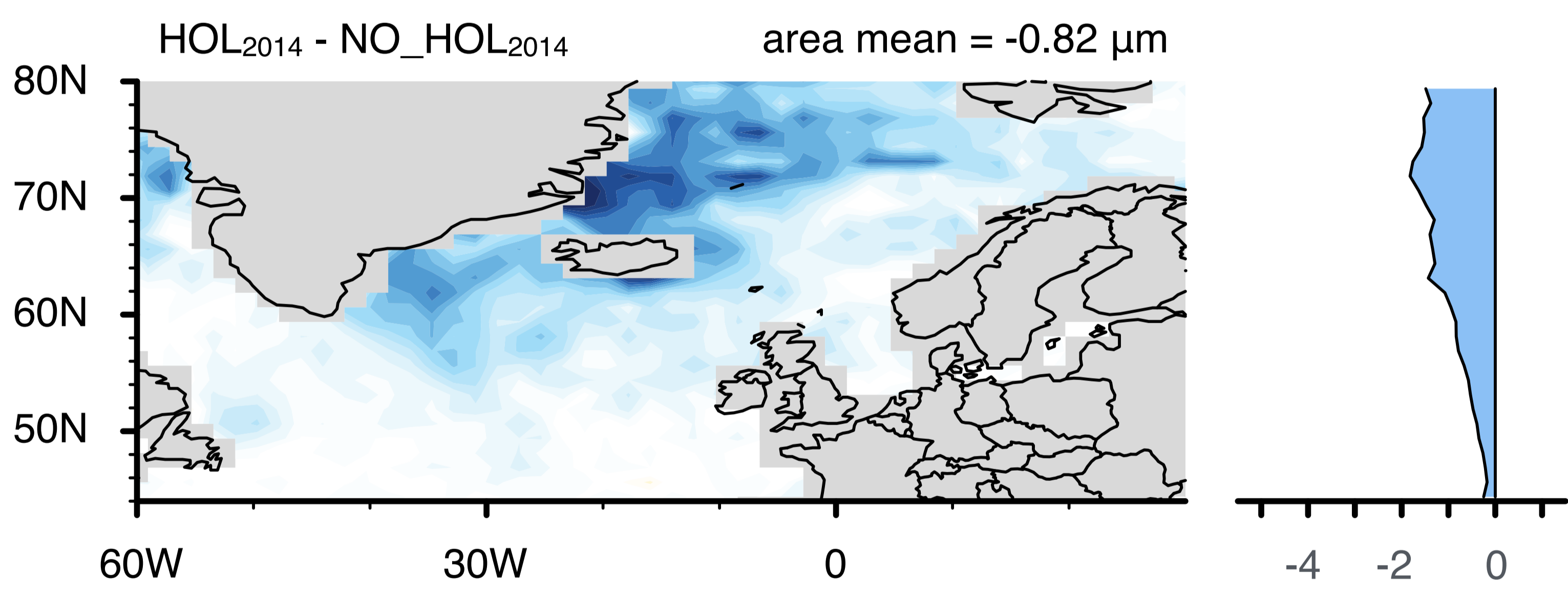
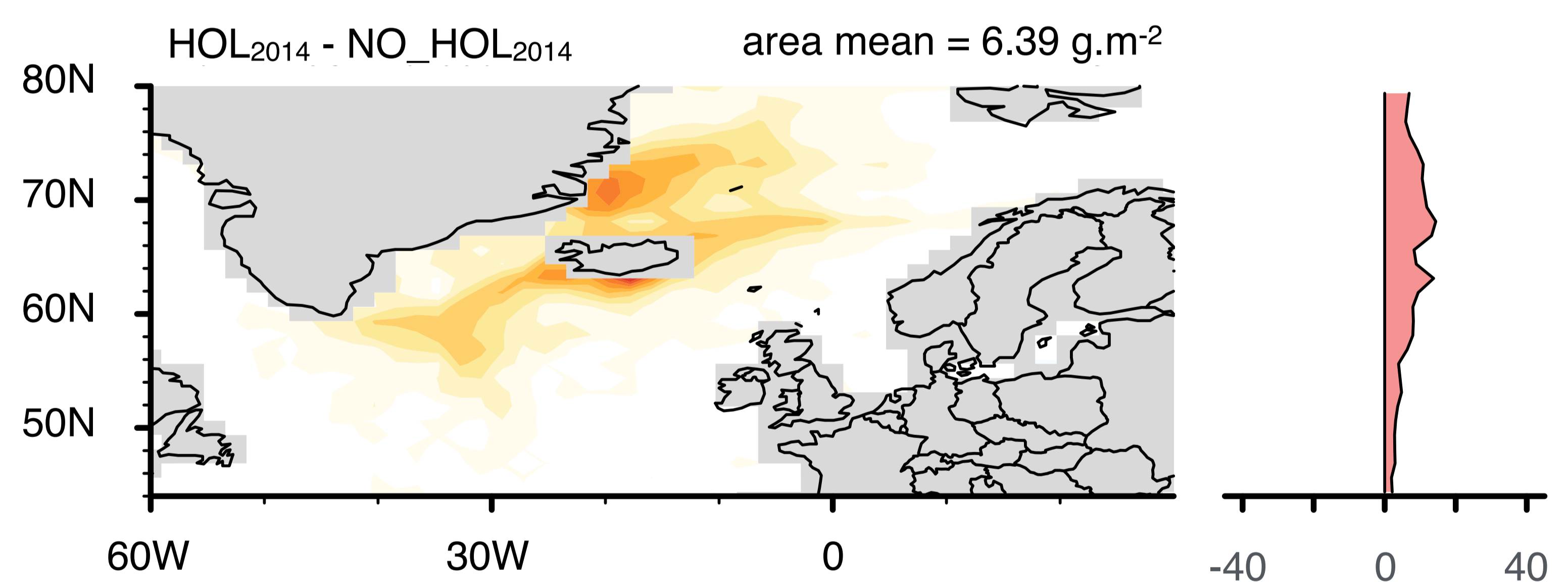
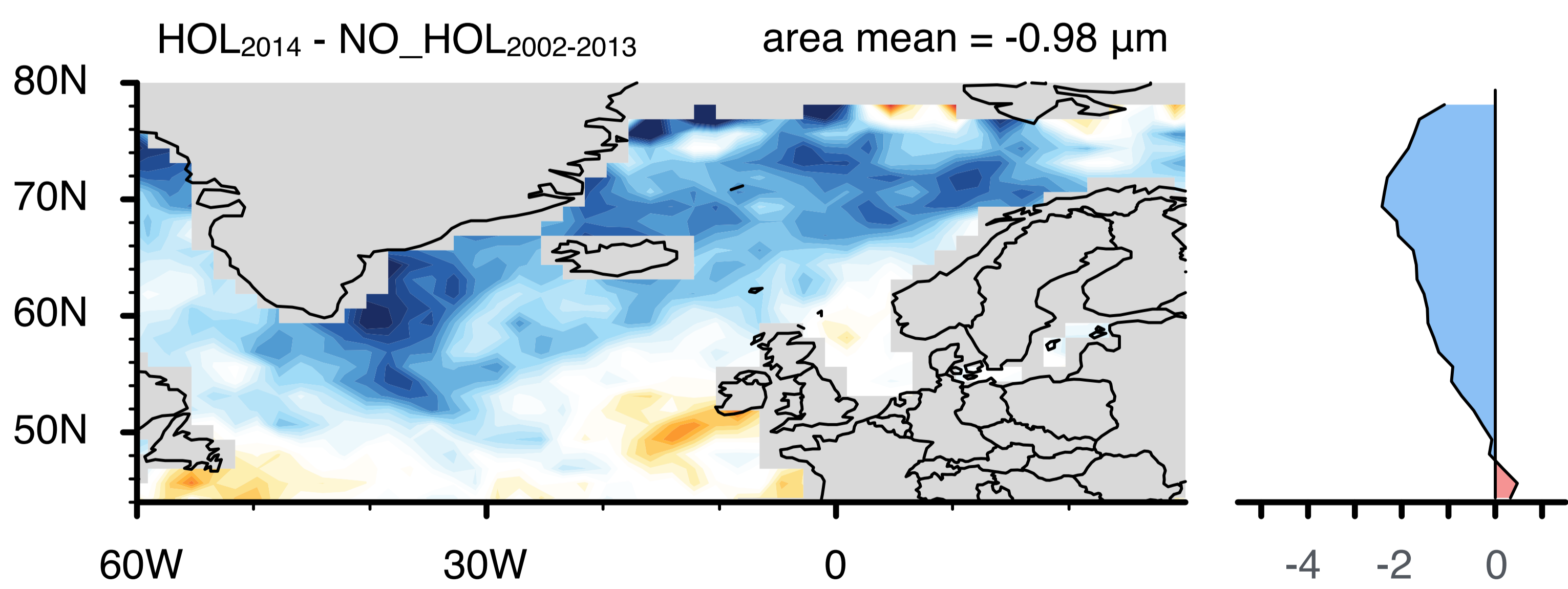
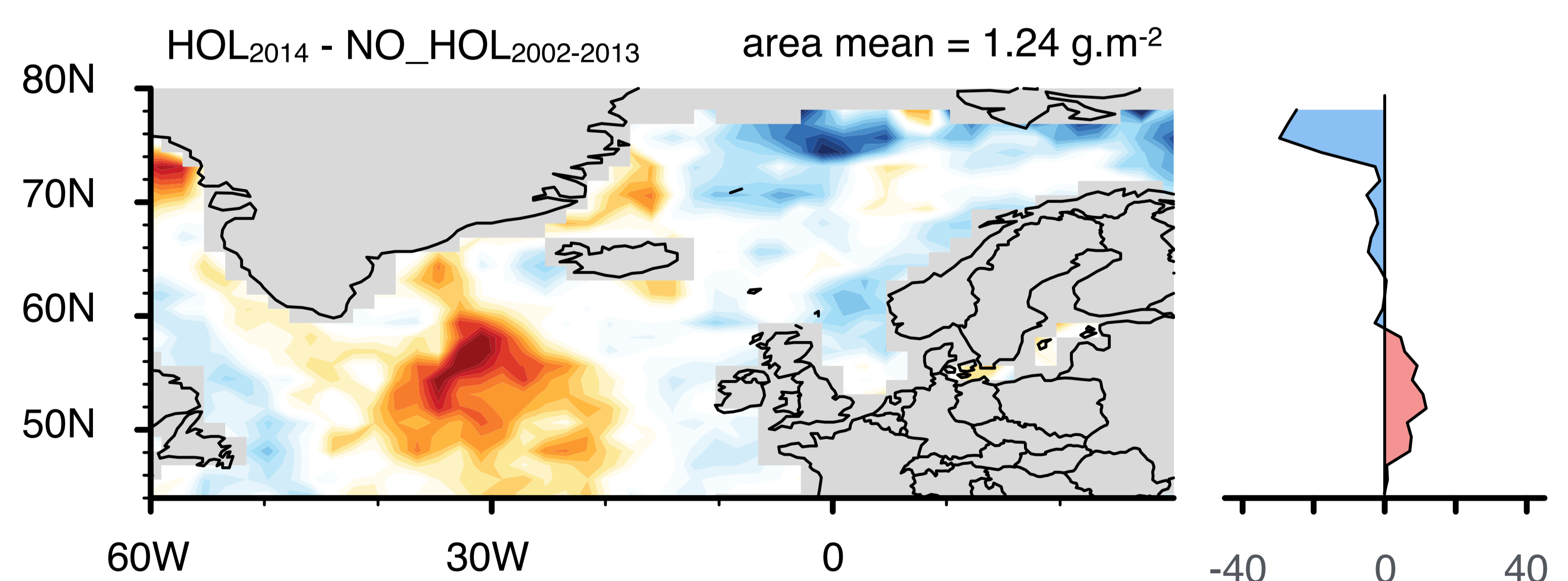
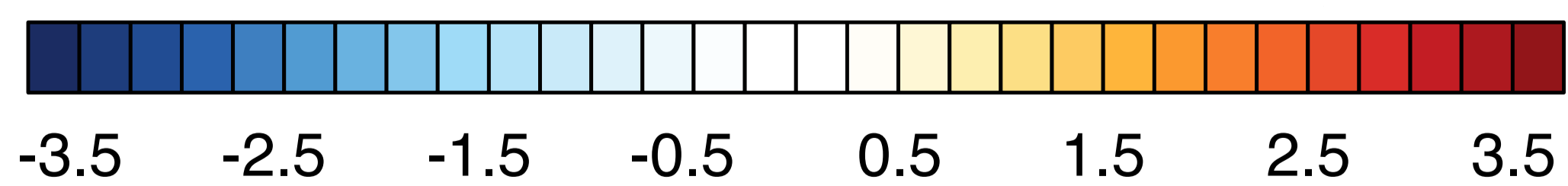
c



d



a ΔAOD **b** ΔN_d **c** Δr_{eff} **d** ΔLWP **e** $\Delta \tau_{cloud}$ **f** Δ ToA net SW radiation

HadGEM3-UKCA**HadGEM3-UKCA****HadGEM3-CLASSIC****HadGEM3-CLASSIC****CAM5-NCAR****CAM5-NCAR****CAM5-Oslo****CAM5-Oslo****MODIS AQUA C051****MODIS AQUA C051**Liquid cloud effective radius anomalies, Δr_{eff} [μm]Liquid Water Path anomalies, ΔLWP [$\text{g}\cdot\text{m}^{-2}$]