

# On the feasibility of monitoring carbon monoxide in the lower troposphere from a constellation of Northern Hemisphere geostationary satellites: global scale assimilation experiments (Part II)

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## Abstract

This paper describes the second phase of an Observing System Simulation Experiment (OSSE) that utilizes the synthetic measurements from a constellation of satellites measuring atmospheric composition from geostationary (GEO) Earth orbit presented in part I of the study. Our OSSE is focused on carbon monoxide observations over North America, East Asia and Europe where most of the anthropogenic sources are located. Here we assess the impact of a potential GEO constellation on constraining northern hemisphere (NH) carbon monoxide (CO) using data assimilation. We show how cloud cover affects the GEO constellation data density with the largest cloud cover (i.e., lowest data density) occurring during Asian summer. We compare the modeled state of the atmosphere (Control Run), before CO data assimilation, with the known “true” state of the atmosphere (Nature Run) and show that our setup provides realistic atmospheric CO fields and emission budgets. Overall, the Control Run underestimates CO concentrations in the northern hemisphere, especially in areas close to CO sources. Assimilation experiments show that constraining CO close to the main anthropogenic sources significantly reduces errors in NH CO compared to the Control Run. We assess the changes in error reduction when only single satellite instruments are available as compared to the full constellation. We find large differences in how measurements for each continental scale observation system affect the hemispherical improvement in long-range transport patterns, especially due to seasonal cloud cover. A GEO constellation will provide the most efficient constraint on NH CO during winter when CO lifetime is longer and increments from data assimilation associated with source regions are advected further around the globe.

## 44 1. Introduction

45

46

47 Observing system simulation experiments (OSSEs) are a powerful method for  
48 evaluating the impact of potential future observations (Edwards et al., 2009;  
49 Timmermans et al., 2015). In Barré et al., 2015a (hereafter, Part I), we introduced  
50 the OSSE framework and method to simulate observations for a future constellation  
51 of geostationary (GEO) satellites. The OSSE results presented in this second part of  
52 the study focus on assimilation of the simulated carbon monoxide (CO) observations  
53 and evaluation of the impact on chemical weather prediction in the northern  
54 hemisphere (NH) troposphere. Because CO is a primary pollutant, with significant  
55 sources from industrial and urban fossil/biofuel burning, wildfires and biomass  
56 burning, it is a convenient chemical tracer for assessing the utility of assimilated  
57 GEO measurements for quantifying pollution emissions and their subsequent  
58 transport.

58

59 We observe high CO concentrations in the lower troposphere and in the NH  
60 due to urban and industrial pollution, over East China, India, Western Europe and  
61 the United States. Other major sources of CO in the NH are wildfires that occur  
62 during dry seasons; e.g., May to October in extratropical northern latitudes,  
63 especially in forested boreal regions. CO is also a reactive chemical compound with  
64 chemical production and destruction mainly due to hydrocarbon and hydroxyl  
65 radical (OH) oxidation, respectively. OH availability governs CO lifetime, which is  
66 shorter during summer and over low latitudes, and longer during winter and over  
67 high latitudes in the NH (Edwards et al., 2004). Satellite instruments can observe CO  
68 plumes from strong emission sources on global scales, travelling distances that  
69 depend on CO lifetime (weeks to months). This makes CO an excellent candidate for  
70 tracking fossil fuel and biomass burning emissions as they are transported from the  
71 sources into the global troposphere. The OH seasonal cycle leads to a CO build-up at  
72 the end of the NH winter, commonly underestimated in model simulations (e.g.,  
73 Stein et al. 2014). This bias is likely due to a combination of factors, including an  
74 underestimation of the magnitude of the emissions, biases in the OH fields, as well  
75 as transport errors (e.g., Jiang et al., 2013; Strode et al. 2015). Data assimilation of  
76 CO, i.e. representing the best CO estimate of the atmosphere using models and  
77 observations, has many applications ranging from air quality characterization,  
78 emissions estimation, large-scale pollutant transport, and climate evolution due to  
79 changing atmospheric composition.

79

80 Part I of this study demonstrated the feasibility of simulating CO  
81 observations from three instruments with characteristics similar to the  
82 Measurement of Pollution in The Troposphere (MOPITT) instrument flying on the  
83 NASA Terra satellite. These three CO instruments would cover the most populated  
84 and hence most polluted areas of the world: Continental US (CONUS), Western  
85 Europe and Eastern Asia. Measurement simulations provide realistic multispectral  
86 sensitivities peaking at the surface during daytime for land observations. These  
87 simulated measurements also provide errors and cloud coverage at variable  
88 horizontal resolution for assimilation experiments. Previous OSSE studies assessed  
89 the impact of GEO instrument capabilities using data assimilation, but with a focus  
90 on the regional scale. Edwards et al., 2009 and Zoogman et al., 2011, 2014a,b  
91 focused on CONUS CO and ozone (O<sub>3</sub>), while Claeys et al., 2011 assimilated GEO

91 measurements of CO and O<sub>3</sub> over Europe and Yumimoto, 2013 assimilated GEO  
92 measurements of CO over East Asia.

93 In part II of this study, we assimilate simulated observations from a GEO  
94 constellation composed of the three instruments defined in Part I into a global  
95 chemistry model to assess the global-scale impacts of GEO satellites for the first  
96 time. The focus of this paper is to quantify the potential of a GEO constellation for  
97 constraining NH CO distributions, especially in the lower troposphere near  
98 anthropogenic sources. We present results from two data assimilation experiments  
99 during summer and winter. Observations from each GEO instrument are assimilated  
100 independently and jointly to evaluate the impact of observations in each domain as  
101 compared to the full constellation

102 Section 2 of this paper further describes the OSSE framework introduced in  
103 Part I, with details on the nature run and the control run. We briefly summarize the  
104 observation simulations covered in detail in Part I, focusing here on cloud cover  
105 variability over different regions and different seasons. We also present the data  
106 assimilation methodology following Barré et al., 2015b. Section 3 gives a detailed  
107 evaluation of the GEO constellation performance due to each instrument with  
108 assimilation results such as increments, global impact on CO errors and skill score  
109 metrics. Section 4 concludes with seasonal and geographical observational  
110 requirements for a GEO constellation of CO measurements and perspectives on  
111 future work using our OSSE framework.

112

## 113 **2. OSSE Setup**

114

115 An OSSE comprises several elements (see part I, figure 1): a Nature Run (NR)  
116 that represents the atmospheric true state; an observation simulator that samples  
117 the NR to produce synthetic observations; a Control Run (CR) that is the modeled  
118 state of the atmosphere; and an assimilation system that merges the synthetic  
119 observations with the CR to produce an Assimilation Run (AR). By comparing the  
120 NR, CR and AR one can assess the impact of a new instrument concept, in this case a  
121 constellation of GEO satellites over the NH.

122

### 123 **2.1 Nature run and control run description**

124

125 The GEOS5 Nature Run (NR) used for simulating the GEO constellation  
126 observations is described in Part I of this paper and complete details and validation  
127 documents are available at <http://gmao.gsfc.nasa.gov/projects/G5NR/>. In this  
128 section, we focus on how we model and parameterize the NR CO concentrations and  
129 emissions. For this study, we use reduced horizontal resolution ( $0.5^\circ \times 0.5^\circ$ ) derived  
130 from the high horizontal resolution run ( $0.06^\circ \times 0.06^\circ$ ) that simulates year 2006  
131 atmospheric conditions. The NR uses a simplified version of CO chemistry as  
132 described in Ott et al., 2010. The only sink for CO is the reaction with OH.  
133 Tropospheric OH is parameterized using OH monthly means from previous  
134 calculations (also for year 2006) of a full chemical mechanism (Duncan et al., 2000).  
135 It was necessary to increase the CO emissions from fossil fuels, biofuels and biomass  
136 burning by 20%, 19% and 11%, respectively, to account for CO production from  
137 non-methane hydrocarbons emitted from these sources. We use monthly mean

138 methane fields to calculate CO produced by methane oxidation as described in Bian  
139 et al., 2007.

140 Detailed descriptions of emissions are provided in Putman et al., 2014.  
141 Biogenic and methane sources of CO are taken from a coarse resolution ( $4^\circ \times 5^\circ$ )  
142 chemical transport model simulations, while biomass burning and fossil fuel  
143 emissions were produced at  $0.1^\circ$  to introduce spatial heterogeneity into the  
144 simulations. We obtain daily CO biomass burning emissions from the Quick Fire  
145 Emissions Dataset (QFED) version 2.4-r6 and CO anthropogenic emissions are  
146 mainly from the Emissions Database for Global Atmospheric Research (EDGAR). We  
147 have disaggregated these emissions in time (yearly to monthly time scales) using  
148 information on the seasonal cycle of fossil fuel emissions from Bey et al., 2001. We  
149 apply no diurnal or weekly variation to the EDGAR emission inventory.

150 We evaluate NR mixing ratios using a combination of surface and satellite  
151 observations. In general, the NR tends to underestimate CO mixing ratios, especially  
152 during extratropical NH spring. We improve significantly these underestimates  
153 through application of an empirically derived bias correction method as described  
154 in <http://gmao.gsfc.nasa.gov/projects/G5NR/TM2014-104606v36.pdf>, leading to a  
155 reduced overall bias of 10% at NH extratropical latitudes compared to MOPITT CO  
156 observations. The NR succeeds in capturing major CO features due to fossil fuel  
157 emissions and biomass burning that are seen in the observations.

158 We use the Community Atmospheric Model with Chemistry (CAM-chem)  
159 version 5 with on-line meteorology (using CAM5 physics, Conley et al. (2012)) and  
160 on-line full gas phase chemical mechanism (MOZART-4 tropospheric chemistry) as  
161 the Control Run (CR) and as a basis for the Assimilation Runs (AR). In this study, we  
162 use a horizontal resolution of ( $1.25^\circ$  longitude by  $0.9^\circ$  latitude) with 30 vertical  
163 levels from the surface up to 4hPa. Emmons et al. 2010, describes and evaluates the  
164 MOZART-4 chemical scheme; Lamarque et al. (2012) and Tilmes et al. (2015)  
165 describe updates to this scheme. The tropospheric version of the MOZART  
166 mechanism includes 85 gas-phase species, 12 bulk aerosol compounds, 39  
167 photolysis and 157 gas-phase reactions. We prescribe the relevant chemical  
168 variables in the stratosphere, between 50 hPa and the top of the model, using  
169 climatology. Lamarque et al. (2012), Tilmes et al. (2015) and Barré et al. (2015b)  
170 showed that the modeled CO distribution at high NH latitudes is underestimated by  
171 values ranging from 25% to 75% when compared to surface, aircraft and satellite  
172 observations, indicating an underestimation of CO emissions and possibly also an  
173 overestimate of the CO loss by OH. Barré et al. (2015b) showed that assimilation of  
174 infrared Low Earth Orbiter (LEO) sounder observations partially or totally corrects  
175 the CR CO bias depending on sounder spatial coverage and vertical sensitivity.

176 We base the CAM-Chem anthropogenic emissions on the Atmospheric  
177 Chemistry and Climate Model Intercomparison Project (ACCMIP) historical  
178 emissions (1960-2000) and RCP 8.5 future scenario emissions (Lamarque et al.,  
179 2010). We use biomass-burning emissions provided by the Fire Inventory from  
180 NCAR version 1.5 (FINNv1.5; Wiedinmyer et al., 2011). We generate biogenic  
181 emissions offline using the global Model of Emissions of Gases and Aerosols from  
182 Nature (MEGAN v2.1; Guenther et al., 2012) and use monthly averages of daily  
183 emissions from MEGAN and FINN emitted at the surface level. Using a monthly  
184 average for the fire emission inventory is a likely source of error given that fires

185 have daily evolving signatures. However, monthly emissions are justified in a global  
186 scale approach with coarse horizontal resolution where large-scale fire signatures  
187 last several months.

188 Figure 1. shows the seasonal tropospheric CO averages from the NR and CR  
189 for the winter and summer cases. Compared to the NR, the CR underestimates the  
190 CO field by 20% to 30% at extratropical NH latitudes. Underestimates of this  
191 magnitude are common in CO simulations as demonstrated by Shindell et al. (2006)  
192 who compared CO fields generated by 26 chemical transport models. As stated  
193 above, CO is primarily emitted in the troposphere from anthropogenic and biomass  
194 burning emissions. However, a significant fraction of tropospheric CO is produced  
195 from chemical oxidation and removed through its reaction with OH. Figure 1. also  
196 shows the correlation coefficients between the NR and CR for the two seasons of  
197 interest. Correlation coefficients range from 0.3 to 0.8 depending on the season and  
198 regions in the northern hemisphere. These values are also in the range of what has  
199 been previously shown by Shindell et al. (2006), Table 4, which gives correlations  
200 ranging from 0.3 to 0.9 for comparisons of chemical transport models with MOPITT  
201 CO data. Overall, we find the CR errors to be realistic in terms of bias and variability.

202 Figure 2. shows emission budgets over the three regions of interest (fields of  
203 regard of the 3 GEO instruments, see Part I figure 4): North America, Western  
204 Europe and Eastern Asia. We display the total emissions (anthropogenic + biomass-  
205 burning + biogenic) and the biomass-burning fraction. The differences between NR  
206 and CR emissions budgets are representative of current model capabilities since  
207 fossil fuel emissions inventories are mostly underestimated (Shindell et al., 2006).  
208 Limitations in state-of-the-art models lead to large uncertainties when  
209 characterizing biomass-burning emissions from fire events (Wiedinmyer et al.,  
210 2011) and hence large differences between the CR and NR. In most cases, there is an  
211 underestimation of CR emissions compared to the NR (for both total emissions and  
212 biomass burning emissions), except for Eastern Asia where very intense fires take  
213 place over Northern Thailand, Myanmar and Laos in NH spring. For South East Asian  
214 fires, the CR largely overestimates the fire emissions compared to the NR. This is  
215 reflected in the emission budgets in Figure 2., i.e., the March budget over Asia. In our  
216 OSSE framework, this fire occurrence over Asia provides a case study that allows  
217 assessment of how well GEO satellite data assimilation constrains the atmospheric  
218 CO state under a change of sign in the emission bias.

219 In summary, differences between the NR and CR are within the range of  
220 differences between state-of-the-art models and observations.

221

## 222 **2.2 Simulated CO observations**

223

224 Part I of this study provided a full description of the synthetic observations  
225 simulated from the NR and showed the instrument footprints, sensitivity and errors,  
226 and impacts of cloud cover on pixel resolution. Part I focused on July 2006 and  
227 described the three instruments that are envisioned: GEO-US (North America),  
228 GEO-EU (Western Europe) and GEO-AS (Eastern Asia). The reader should refer to  
229 Part I for more details about the observation simulations. In this Part II paper, we  
230 extend the observation simulation data set to January, February, March (JFM) and  
231 June, July, August (JJA) 2006.

232 Cloud cover is important as it limits the capability of a remote sensing  
233 instrument to monitor tropospheric composition. Figure 3 displays the three  
234 instrument footprints and the cloud free ratio for JFM and JJA, 2006. The cloud free  
235 ratio is the number of cloud free occurrences over the total number of possible  
236 measurements for a given pixel. Over the three observational domains, differences  
237 of cloud free ratio between winter and summer are large. Europe and North America  
238 show more data coverage during summer than winter. This tendency is reversed for  
239 Asia. Over extratropical latitudes, summer is generally significantly less cloudy than  
240 winter due to warmer air that can retain more water vapor. Over Asia, the GEO  
241 instrument field of view (see part I figure 4) tends to cover tropical and subtropical  
242 regions, and is subject to the Asian monsoon during summer, which is a relatively  
243 wet season. For GEO-AS, winter is drier than summer with fewer clouds and more  
244 data coverage.

245 The geographical structure of data coverage changes with season and  
246 exhibits complex patterns. GEO-EU shows a North-South coverage difference with  
247 high coverage at southern latitudes and almost no observations northward of 50°N  
248 during the winter. Good coverage over the Mediterranean is even higher during  
249 summer. GEO-US shows very low winter coverage over New England and the Great  
250 Lakes area but reasonable coverage (above 30%) elsewhere. Summer provides  
251 overall good coverage (above 30%) and excellent coverage (above 80%) over  
252 California. GEO-AS shows patterns that are more complex, e.g., very high coverage  
253 and very low coverage over the southwest part of the domain and over the Japanese  
254 east coast, a North-South coverage difference that is less marked over winter than in  
255 summer. Overall, land data coverage is higher in winter than in summer over Asia.

256

## 257 **2.3 Assimilation system and global OSSE design**

258

### 259 **2.3.1 Synthetic meteorological observations**

260

261 In this OSSE framework we use the Data Assimilation Research Testbed  
262 (DART, Anderson et al. 2009), which is a community data assimilation software  
263 package developed since 2002 at the National Center for Atmospheric Research  
264 (NCAR). DART implements the Ensemble Kalman Filter (EnKF) technique originally  
265 introduced by Evensen (1994). This software is designed to provide high modularity  
266 that allows an easy interface for a variety of models. It facilitates ensemble-based  
267 data assimilation (DA) without needing to construct a model adjoint and adjoints for  
268 observation operators as in the case of 4D variational-based DA.

269 DART assimilates meteorological and chemical observations simultaneously.  
270 The data assimilation setup is based on the work of Raeder et al. (2012) for the  
271 meteorology assimilation and Barré et al. (2015b) (see supplementary information  
272 document) for the chemistry-meteorology assimilation, where conventional  
273 NOAA/NCEP meteorological observations are assimilated. These two studies  
274 provide a detailed evaluation of the performance of the meteorological analysis  
275 produced with the DART setup. In this present study, we generate synthetic  
276 conventional meteorological observations by sampling the nature run variables  
277 (winds, temperature and specific humidity) at real observation locations. We define  
278 the error characterization of synthetic observations using the ratio of the real

279 observation error over the measurement value. We then add random noise to the  
 280 sampled nature run values according to the specified error of the synthetic  
 281 observations. We use the following relationships to generate synthetic  
 282 meteorological observations:

$$283$$

$$284 \quad X_s = X_t + \varepsilon \quad (1)$$

$$285$$

$$286 \quad \varepsilon = \mathcal{N}(0,1) \cdot e_s \quad (2)$$

$$287$$

$$288 \quad e_s = X_t \cdot e_m \cdot X_m^{-1} \quad (3)$$

289  
 290 Where  $X_s$  is the synthetic observation value,  $X_t$  the nature run sampled at the real  
 291 observation location,  $\varepsilon$  the measurement noise,  $e_s$  the synthetic observation error,  $e_m$   
 292 the real observation error and  $X_m$  the real observation value and  $\mathcal{N}(0,1)$  a standard  
 293 normal distribution. For meridional (V) and zonal (U) wind simulated  
 294 measurements, we take into account the wind speed ( $\sqrt{U^2 + V^2}$ ), to avoid infinite or  
 295 very large ratios when calculating the ratio  $X_t / X_m$  in equation 3. In the OSSE  
 296 experiments, the CR assimilates only meteorological data, while the ARs assimilate  
 297 both meteorological and CO data. The following section describes the experimental  
 298 design of this study.

### 300 **2.3.2 GEO constellation CO assimilation experimental design**

301  
 302 Barré et al. (2015b) provides a complete description of the chemical data  
 303 assimilation setup with a focus on CO and shows the results and evaluation with  
 304 independent measurements from assimilating the MOPITT and the Infrared  
 305 Atmospheric Sounding Interferometer (IASI) instrument retrieved CO profiles into  
 306 the CAM-Chem model. That paper highlights the different capabilities of the IASI and  
 307 MOPITT instruments with particular attention to instrument vertical sensitivity and  
 308 coverage and their impact on the analysis of global CO atmospheric composition.  
 309 Barré et al. (2015b) showed that satellite observations that have frequent revisit  
 310 and enhanced vertical sensitivity toward the surface close to sources provide an  
 311 efficient constraint and generate a global improvement in tropospheric CO  
 312 concentrations. In the present study, we use the same MOPITT CO data assimilation  
 313 setup to assimilate a geostationary constellation of simulated MOPITT-like  
 314 measurements. Although it is possible to infer changes in the concentrations of  
 315 other chemical species, here we only adjust CO concentrations using data  
 316 assimilation of CO observations, as in Barré et al. (2015b).

317 We assimilate the full GEO constellation and each instrument independently in  
 318 order to assess the global impact of the constellation and understand the  
 319 contribution of each instrument to the estimation of the NH CO field. These  
 320 assimilation experiments are repeated over the winter and summer 2006 (January-  
 321 February-March and June-July-August, respectively) because emissions, cloud cover  
 322 and CO chemical lifetime change significantly throughout the year. We hereafter  
 323 name the different assimilation runs as follows:

- 324 • Control run (CR): we assimilate only meteorological data;

- 325 • Full constellation assimilation run (AR0): we assimilate meteorological and 3  
326 GEO instrument data;
- 327 • GEO-US assimilation run (AR1): we assimilate meteorological and US GEO  
328 instrument data;
- 329 • GEO-EU assimilation run (AR2): we assimilate meteorological and European  
330 GEO instrument data;
- 331 • GEO-AS assimilation run (AR3): we assimilate meteorological and Asian GEO  
332 instrument data.

333

### 334 **3. Results**

335

#### 336 **3.1 Data assimilation increments**

337

338 In this section, we investigate the overall constraint on model CO fields from  
339 the AR0 assimilation experiment during winter and summer. Figure 4 displays the  
340 root-mean-square (RMS) of the relative increments (posterior minus prior  
341 normalized by the prior) over a month for the 6-hourly data assimilation window.  
342 As described in Barré et al. (2015b) CO retrievals are assimilated every 6 hours and  
343 the RMS of the relative increments over a month is useful for identifying the overall  
344 magnitude of the CO changes due to assimilation, and for detecting short-term  
345 systematic error patterns in the CR. Please refer to Barré et al. (2015b) for  
346 additional details about the data assimilation setup.

347

348 We can observe seasonal differences in the magnitude of the increments.  
349 Three main factors can explain this difference: cloud coverage, CO model error and  
350 hence CO emissions error, and instrument sensitivity. During winter over Europe  
351 and North America, relative increments are smaller than during summer because we  
352 assimilate less data due to higher cloud cover. Conversely, Asia has the opposite  
353 tendency with relative increments that are larger over winter due to less cloud  
354 cover (see section 2.2 and Figure 3). In general, errors in CO emissions tend to be  
355 larger during the summer than during the winter (Figure 2). This also explains  
356 larger increments during the summer. Confirmation of this comes from relative  
357 increments showing structural patterns related to emission patterns. For example,  
358 we observe large relative increments over the Northeast United States (New  
359 England and slightly lower latitudes) where there are large anthropogenic CO  
360 emissions throughout the year due to high urbanization in this area. We also  
361 observe large relative increment patterns around the Bohai Sea (near Beijing)  
362 where urbanization is very high as well.

362

363 We also capture fire structures in the data assimilation relative increments;  
364 these are visible over South East Asia during winter where we detect very strong  
365 fire occurrences. Emission budgets in Figure 2 show that the CR overestimates this  
366 fire source compared to the NR. We detect other fire patterns over North America  
367 and Europe during summer, e.g., Central North US, North West US and Spain. We can  
368 also explain relative increment magnitudes in Figure 2 from differences between the  
369 CR and NR emission budgets. If the differences in the emission budget are large in a  
370 given region, then the magnitude of the data assimilation relative increments is also  
likely to be large.



371 We note that instrument sensitivity is the least dominant factor in relative  
372 increment size. We calculated the seasonal average degrees of freedom for signal  
373 (DFS), which represents the independent vertical information in the measurement  
374 throughout the troposphere, (see part I for details). GEO-US shows a DFS of 1.53  
375 (1.28) during the winter (summer), GEO-EU shows a DFS of 1.40 (1.30) during the  
376 winter (summer) and GEO-AS shows a DFS of 1.61 (1.36) during the winter  
377 (summer). DFS depends primarily on thermal contrast and CO abundance and for  
378 these observational domains, there is clearly weaker instrument sensitivity during  
379 the summer. However, the seasonal differences in sensitivity are not so large that  
380 they dominate the relative size of increments found for summer versus winter.

381 Diagnosing data assimilation relative increments shows that a GEO  
382 constellation provides an efficient constraint on atmospheric CO on continental  
383 scales at or close to the main anthropogenic CO sources over the NH. In the  
384 observing domain for this constellation, we also detect some fire events, but during  
385 summer, several fires occur outside the constellation field of view that would  
386 require other (e.g., LEO) satellites to monitor and hence cannot be constrained using  
387 only GEO data assimilation.

388

### 389 **3.2 Data assimilation impact**

390

391 To assess the impact of assimilating the GEO constellation on global northern  
392 hemisphere CO, we first compare the full constellation assimilation run AR0 and the  
393 CR with the NR. Figures 5 and 6 show monthly averaged differences over the  
394 troposphere (surface to 200 hPa) of CR and AR0 with NR for winter and summer,  
395 respectively. In the same manner, figure 7 shows the same differences over the  
396 lower troposphere (surface to 800 hPa) just for February and July. Those plots show  
397 the overall biases of CR and AR0 versus NR, respectively. The CR runs show larger  
398 and more extended biases during winter than summer in the entire troposphere as  
399 well as in the lower troposphere. Despite stronger differences in emissions during  
400 summer between CR and NR (see figure 2), the shorter CO lifetime during summer  
401 reduces the global tropospheric bias. We can also observe this effect within the  
402 given seasons in figure 5 and 6. The CO lifetime shortens through January to March  
403 (and June to August) giving a reduced CR bias. With a shorter CO lifetime, errors  
404 owing to CO emissions have less persistence over time and propagation throughout  
405 the troposphere is less likely.

406 The AR0 reduces the overall CO bias in the NH troposphere. Figure 7 shows  
407 that a significant error reduction occurs at the lowest level of the atmosphere close  
408 to the sources over the GEO constellation fields of regard (see part I, figure 4 and  
409 figure 3 of this paper). As a result, data assimilation does not improve major error  
410 patterns close to the surface and out of the fields of regard (e.g., CO fire emissions  
411 close to Lake Baikal). A persistent error in the AR0 is still seen with patterns close to  
412 major cities or groups of cities over the 3 regions of interest. This shows that the DA  
413 system used here constrains CO fields close to CO sources, but that this system does  
414 not yet have the capability of updating the CO emission inventory. This means that  
415 while error reduction of the CO fields close to the surface is large, the errors are not  
416 removed since the un-adjusted model CO sources remain as an input to the error in  
417 the atmospheric CO fields. Assimilation of retrieved profiles close to the sources can

418 provide a hemispheric constraint due to long-range transport of the relative  
419 increments and persistence over time of the error correction. In both seasons, global  
420 constraints take about a month for advection to spread the error correction over the  
421 NH. The level of improvement is also dependent on the CR bias. In the winter case  
422 study, the CR bias is larger than in the summer case study leading to the AR0 run  
423 being closer to the NR during summer compared to winter. Even if the error  
424 reduction is global, we observe large errors at the CO source locations because of  
425 remaining biases in emission inventories. For example, over Asia during July  
426 (summer), the cloud cover is high and hence the data density is too low to show  
427 significant improvement of the CO fields close to the surface. This effect is even  
428 more pronounced over the source regions that are not located in the observing  
429 domain of the GEO constellation, e.g. Siberian fires and Canadian fires.

430 Assimilation of a GEO-constellation of CO tropospheric measurement over  
431 the main NH anthropogenic sources allows a partial hemispheric constraint. Section  
432 3.3 will quantify the performance of each satellite instrument.

433

### 434 **3.3 Assimilation performance assessment**

435

436 To quantify the effect of assimilation of the synthetic GEO-constellation  
437 observations, we define the skill score by the following metric:

438

$$439 \quad SkillScore = 1 - \frac{\sum_t (AR - NR)^2}{\sum_t (CR - NR)^2}$$

440

441 This score is the ratio of the square error of the AR with respect to the CR over time  
442  $t$ ; we apply this to every grid cell of the CAM-Chem model. If the skill score is equal  
443 to 1, then the AR is perfect relative to the NR (AR equals NR). A positive value  
444 indicates that the square error of the AR is reduced by the ratio (or percentage)  
445 given by the skill score. If the skill score is zero, then the assimilation provides no  
446 changes; negative values indicate a degradation of the AR compared to the CR.

447 Figures 8 and 9 show the skill scores for the troposphere (surface to 200  
448 hPa) for each month for winter and summer, respectively. We compute skill scores  
449 for the full constellation assimilation AR0, and for the single instrument observation  
450 experiments: AR1, AR2 and AR3. Data assimilation skill scores on single instrument  
451 assimilation (for AR1, AR2 and AR3) demonstrate the time required for a given  
452 instrument assimilation to impact the model tropospheric hemispheric CO. We  
453 identify two main patterns of transport affecting error reduction. The first pattern  
454 involves the Westerlies and warm conveyor belt processes at extratropical latitudes  
455 (AR1, AR2 and AR3). We clearly see this pattern over the first month of assimilation  
456 (January or June) crossing the Atlantic Ocean, the Asian continent and the Pacific  
457 Ocean from East to West. The second pattern involves the trade winds, which  
458 constrain tropical regions (AR1 and AR2 only) as they move from East to West over  
459 the tropical Pacific and the tropical Atlantic. Overall, the skill score shows  
460 improvement for every experiment, but to a different degree. In addition, we can see  
461 a degraded skill score away from the assimilated regions. This can be due to a bias  
462 sign change between the NR and the CR. If the overall assimilation effect is a positive  
463 bias (NR larger than CR) correction but a local negative bias is occurring (NR lower

464 than CR) the assimilation run will show a degraded skill score in that particular case.  
465 Degraded skill scores are also due to coupled meteorology-chemistry processes  
466 represented in the CAM-Chem model. Adjusting the CO in a given region modifies  
467 the tropospheric chemistry budget, which can alter radiatively active species or  
468 provide a feedback on cloud formation and hence modify the meteorology. A  
469 modified meteorology can then affect the chemistry and hence change CO. This  
470 feedback is more obvious over lower latitudes and summer because of more  
471 complex dynamics at lower latitudes and chemistry that is more active during  
472 summer and at lower latitudes.

473 The winter fire event over South East Asia also illustrates these two effects.  
474 In this case, the fire plume is overestimated whereas a global underestimation (bias)  
475 of CO is provided by the CR. Assimilation of remote instruments from Asia will tend  
476 to increase the global CO, but will also contribute to an increase in CO in the fire  
477 plume and hence degrade the skill scores. In addition, high fire emissions generate a  
478 heavily polluted plume over the Pacific. Even slight changes in dynamics can  
479 generate large CO errors if the emission differences between the NR and CR are  
480 large, as it is the case between NR and CR emissions over Asia in March. In Figure 8  
481 during March, the AR1 and AR2 (i.e., GEO-AS not assimilated) shows the signature of  
482 transported errors from the fire plumes, where a pattern of negative skill scores  
483 follows the large fire plume over the Pacific. In AR0 and AR3, where we assimilate  
484 the GEO-AS data, positive values above 0.6 replace the negative skill score pattern.  
485 This shows the importance of constraining the CO fields close to sources to generate  
486 improved remote CO fields, a result that is consistent with the conclusion of Barré et  
487 al. (2015b) using real data from MOPITT.

488 Figures 8 and 9 show large differences in the skill score magnitude over the  
489 NH. During winter, the CO lifetime is more than a factor of 2 longer than over  
490 summer (Shindell et al., 2006 and Edwards et al., 2004) due to oxidant loading  
491 which is greatest during the summer months. CO accumulates more during winter  
492 than during summer, leading to a more negative bias in the CR (see figures 5 and 6).  
493 The CR winter bias is larger than the CR summer bias even though emission  
494 differences are generally smaller during winter (Figure 2). Data assimilation relative  
495 increments, or the error reduction generated by assimilation close to the emission  
496 sources, then have more persistence over time during winter, and are advected  
497 throughout the entire troposphere. The AR0 skill scores show an average maximum  
498 around 0.7 during February 2006 (a month after starting the assimilation) and the  
499 pattern of improvement with respect to NR is relatively homogenous over the entire  
500 NH. During summer, July 2006 shows a 0.7 skill score over assimilated regions  
501 (GEO-US and GEO-EU), but the skill score is lower, down to 0.4, over remote regions.  
502 The reduction in long-range improvement in the AR0 during summer is also due to a  
503 lack of observational constraints over strong boreal fire sources that generate  
504 additional error variability in the CR relative to the NR. By looking at independent  
505 assimilation experiments (AR1, AR2, and AR3), the difference is even more  
506 noticeable.

507 As explained in section 2.2, cloud cover varies from one observed region to  
508 another, and depends on the season. GEO-US and GEO-EU show more data coverage  
509 during summer than during winter, and this tendency is opposite for GEO-AS. From  
510 the skill score seasonal tendency described above, cloud occurrence and hence data

511 coverage is not the dominant factor determining skill scores. During winter, the CO  
512 lifetime is sufficiently long that less data density is sufficient to constrain the  
513 assimilation. Additionally, emission patterns and errors are mostly anthropogenic  
514 and have smaller variability and a more consistent geographical structure over time  
515 compared to fires. During summer, the CO lifetime is shorter and emission patterns  
516 are often more sporadic due to fires. However, during a given season, cloud cover  
517 affects the magnitude of the skill score. Over the GEO-AS footprint, the cloud free  
518 ratio is relatively low during summer (around 20% on average). This leads to lower  
519 skill scores for the summer AR3 experiment. In general, patterns of improvement  
520 are broader in space and larger during winter than summer, despite the reduced  
521 data sampling due to cloud cover over GEO-US and GEO-EU. During winter, the  
522 longer CO lifetime means that assimilating data from a single GEO instrument can  
523 provide a quasi-global improvement, which is not the case for summer.

#### 524 525 526 **4. Conclusions and perspectives**

527  
528 In this second part of our study we assessed the capability of a potential GEO  
529 constellation for monitoring atmospheric composition using an OSSE with a focus on  
530 measurements of CO. Part I of this study demonstrated that 3 GEO instruments  
531 measuring CO from space can be simulated realistically over three major  
532 anthropogenically active regions: CONUS, Western Europe and Eastern Asia. To  
533 perform the OSSE, we assimilated the synthetic constellation measurements into the  
534 CAM-Chem model-using DART. We first assessed differences between the CR and  
535 the NR, and found these to be reasonable based on global model biases, emissions  
536 and CO uncertainties according to literature on state-of-the-art global chemistry  
537 climate models. We designed assimilation experiments to assess the effects of long-  
538 range transport, seasonality, emissions and cloud cover on the capabilities of the  
539 GEO constellation to constrain CO concentrations. We designed two case studies of  
540 3-month assimilation: winter (January-February-March) and summer (June-July-  
541 August). In addition to the control run (meteorological data assimilated only) and  
542 the full constellation assimilation experiment that we use as a benchmark, we also  
543 performed assimilation experiments for each instrument independently. In total, 10  
544 data assimilation experiments led us to the following main conclusions:

- 545  
546 1. Assimilation relative increments (posterior minus prior fields) are mostly  
547 located at or near the emission sources, and through long-range transport,  
548 these impact the entire NH troposphere. This result suggests that model  
549 errors in CO are largely due to emissions, which is consistent with previous  
550 data assimilation and modeling studies (Shindell et al., 2006; Fortems-  
551 Cheiney et al., 2011; Lamarque et al., 2012; Jiang et al., 2013; Barré et al.,  
552 2015b; Inness et al., 2015; Miyazaki et al., 2015; Tilmes et al., 2015). Each  
553 assimilated instrument shows improvement with respect to the CR in the CO  
554 transport patterns over large-scale areas associated with the westerly and  
555 trade winds at different latitudes.
- 556 2. The magnitude of the global impact depends on season. Winter data  
557 assimilation experiments show better improvement in CO NH distributions

558 than for summer. We explain this as follows. First, the CO lifetime during  
559 summer is shorter so that data assimilation relative increments have less  
560 persistence over time and less global advection within the model. Second, the  
561 summer has more large-scale fires in boreal regions, or away from the GEO  
562 constellation fields of regard. These fire emissions that are not captured by  
563 the GEO constellation are important to the global CO budget and variability.  
564 3. Cloud cover affects the quality of the assimilated runs but this effect is not  
565 dominant when comparing summer and winter simulations. Winter shows a  
566 strong decrease of the cloud free ratio (number of cloud free scenes for a  
567 given pixel over a season) compared to summer for GEO-US and GEO-EU.  
568 This tendency is opposite for GEO-AS. However, the magnitude of the  
569 improvement with respect to the CR is still larger during winter due to CO  
570 lifetime, discussed in point 2 above. For summer, GEO-AS provides the lowest  
571 skill scores because of heavy cloud cover due to the Asian monsoon, and  
572 hence weak constraints from simulated CO observations.

573

574 This study assessed the observational requirements for CO, a good indicator of  
575 anthropogenic, fire and other natural emissions that have a lifetime long enough to  
576 allow transport between continents. Requirements are less demanding in terms of  
577 data density during winter compared to summer, and at wintertime extratropical  
578 latitudes compared to the tropics. Over the next decade, instruments will monitor  
579 atmospheric composition from geostationary platforms, (with temporal resolution  
580 on the order of minutes, but with coverage restricted to specific areas), and from  
581 LEO platforms that provide a global picture of the atmosphere but at lower temporal  
582 resolution (a revisit rate of 1 or 2 days). A next step of this study will be to assess  
583 the synergy between GEO and LEO platforms to constrain atmospheric CO  
584 composition and associated emissions from a global perspective. Assimilating the  
585 two different geometries in a single OSSE framework will provide a thorough  
586 scientific assessment.

587 Another focus for future work will be inferring emissions from GEO observations  
588 in order to provide accurate chemical forecasts near the surface. We will use the  
589 OSSE framework as presented here to assess the best method for emission source  
590 inversion using the ensemble Kalman filter (EnKF) technique. This will help define  
591 measurement requirements depending on emission types and their variability (e.g.,  
592 anthropogenic emissions versus biomass burning). We will also investigate a  
593 combined CO and aerosol optical depth (AOD) assimilation with source inversion of  
594 carbonated aerosol species (black carbon and organic carbon).

595

596

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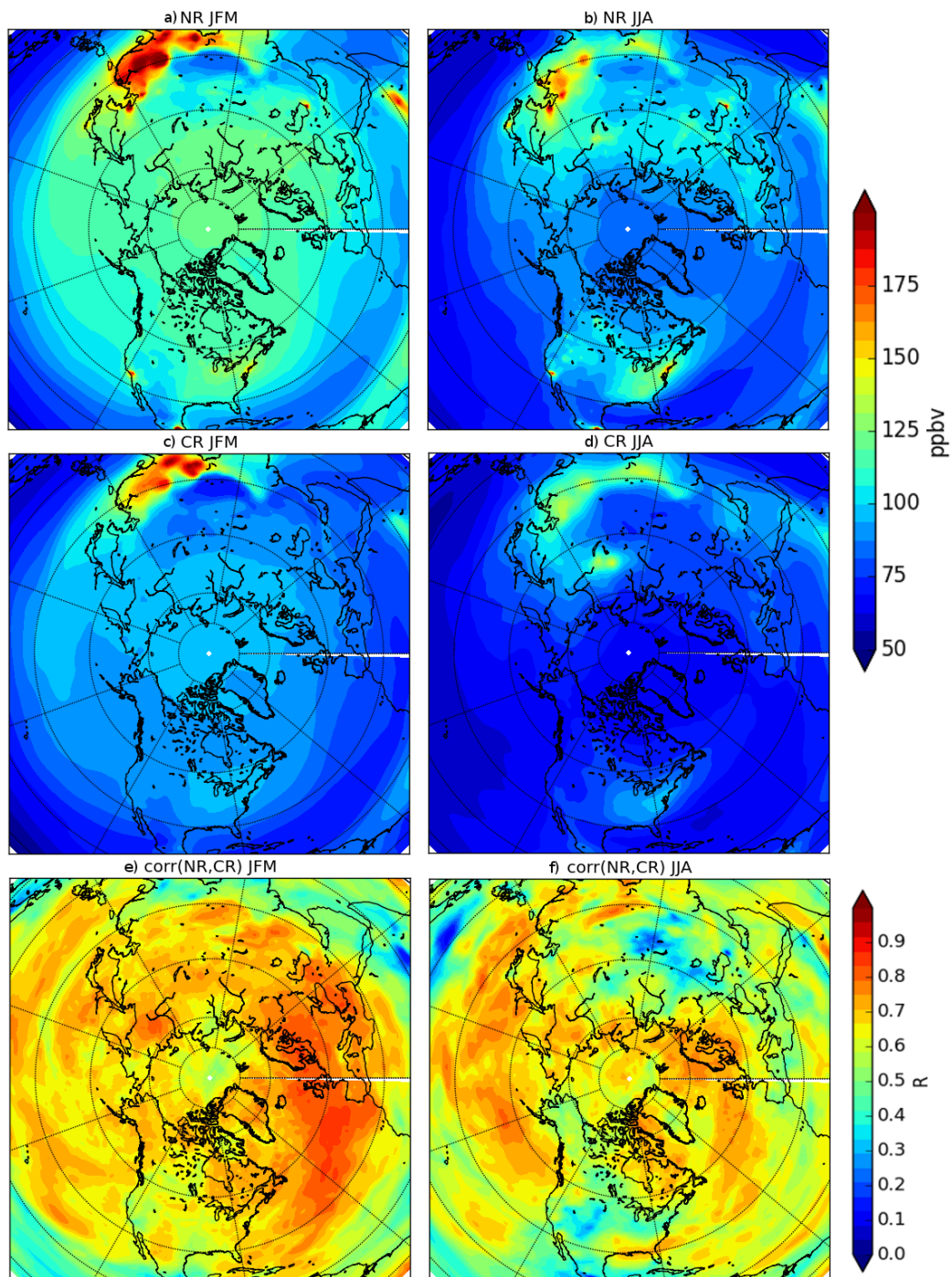
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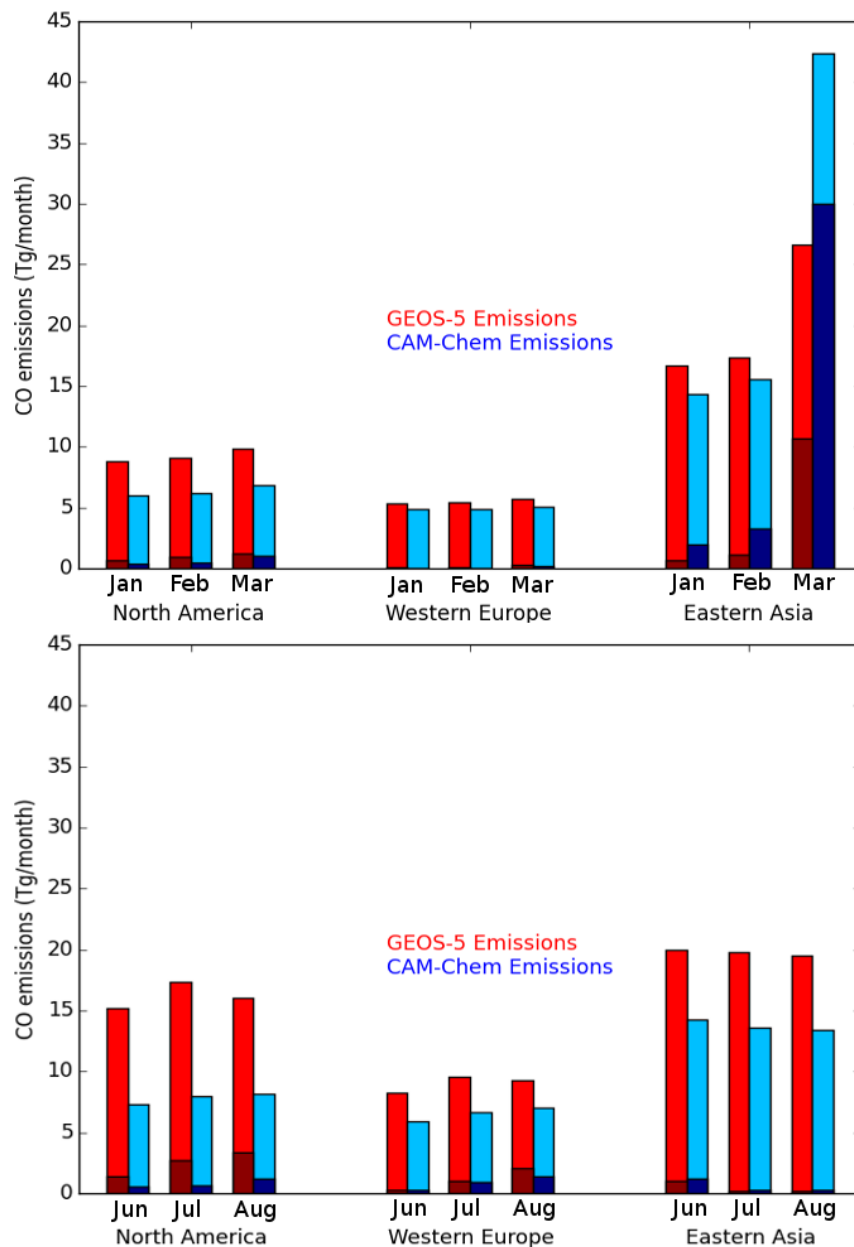
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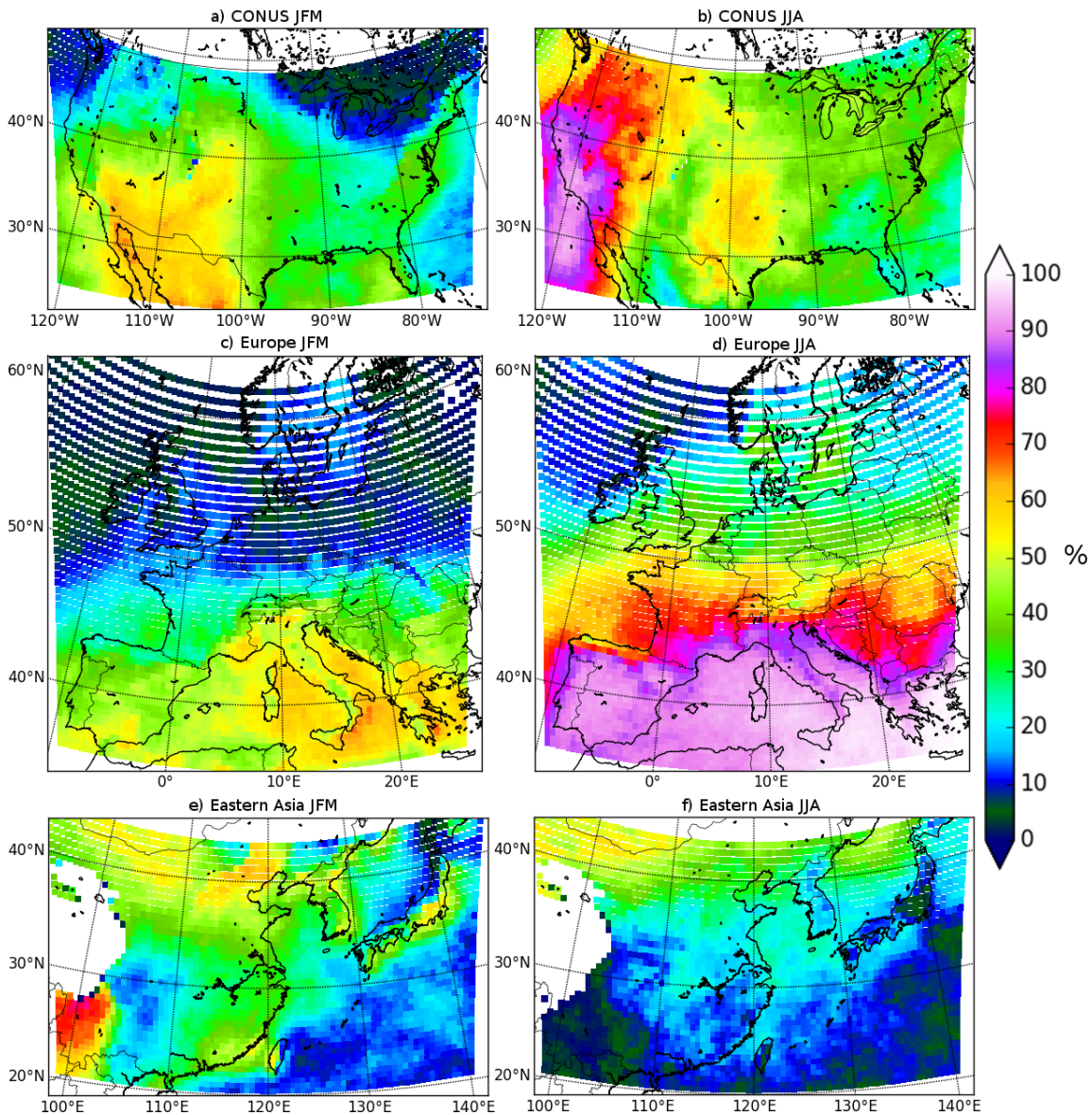
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**Figure 1. Plots of the Nature Run (NR) and the Control Run (CR) January-February-March (JFM) a) and c), respectively, and June-July-August (JJA) b) and d), respectively. We convert mean values of Surface-200hPa tropospheric CO column into a pseudo volume mixing ratio. Red and blue colors refer to relatively high and low values, respectively. Bottom panels show the correlation coefficient  $R$  between the NR and CR for JFM (e) and JJA (f), respectively.**



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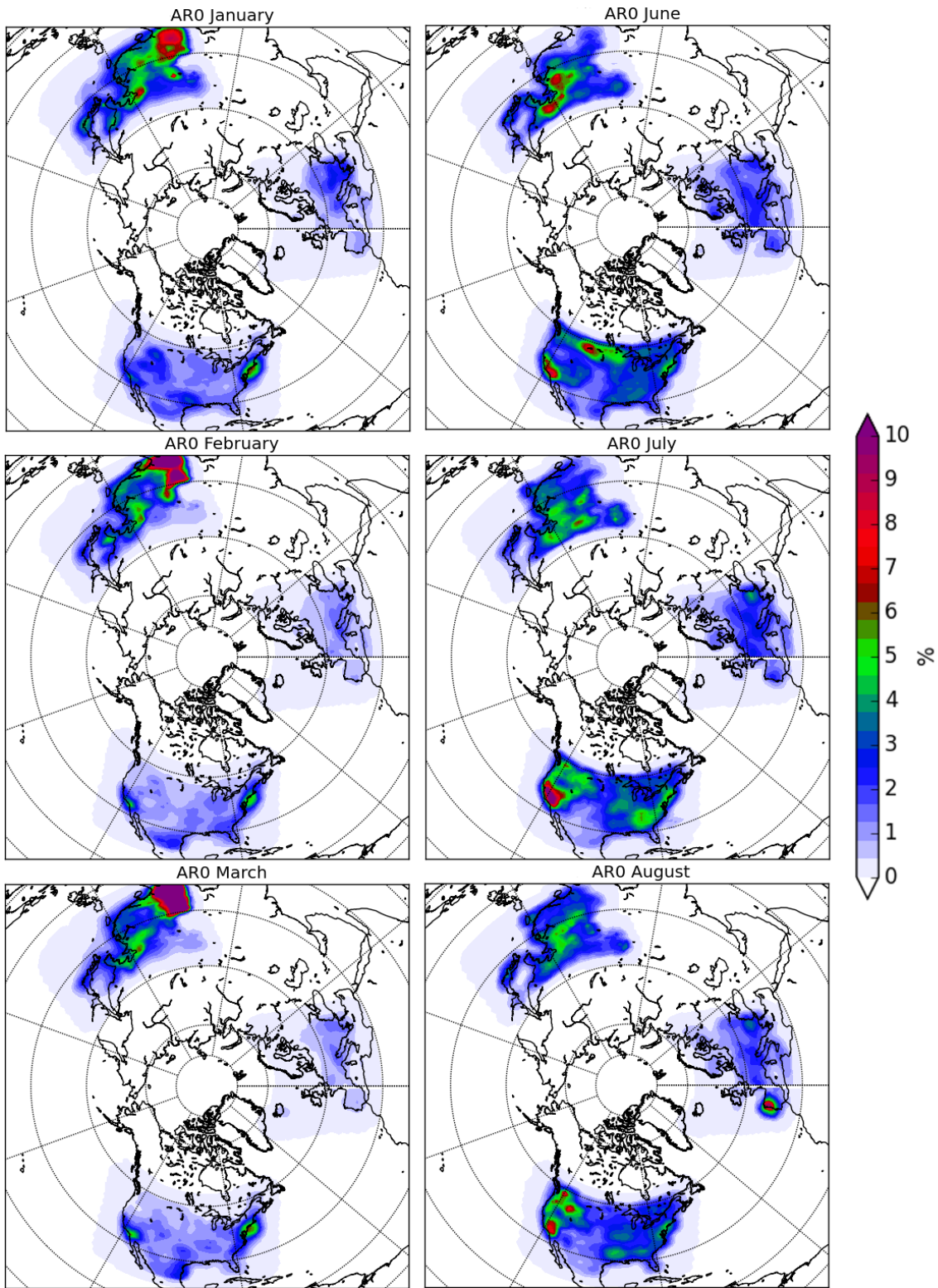
**Figure 2. Monthly emission estimated budgets derived from emission inventories for winter (top panel) and summer (bottom panel) 2006 for GEOS-5 (red) and CAM-Chem (blue) in Teragrams (Tg) of CO per month. Dark colors indicate the biomass-burning fraction of the emission budgets.**



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**Figure 3. Cloud free ratio in % for the three geostationary instruments for the winter (left) and summer (right) seasons. Top panels (a and b) refer to the CONUS; middle panels (c and d) refer to Europe; bottom panels (e and f) refer to Eastern Asia. Red/purple and blue colors refer to relatively high and low values, respectively.**

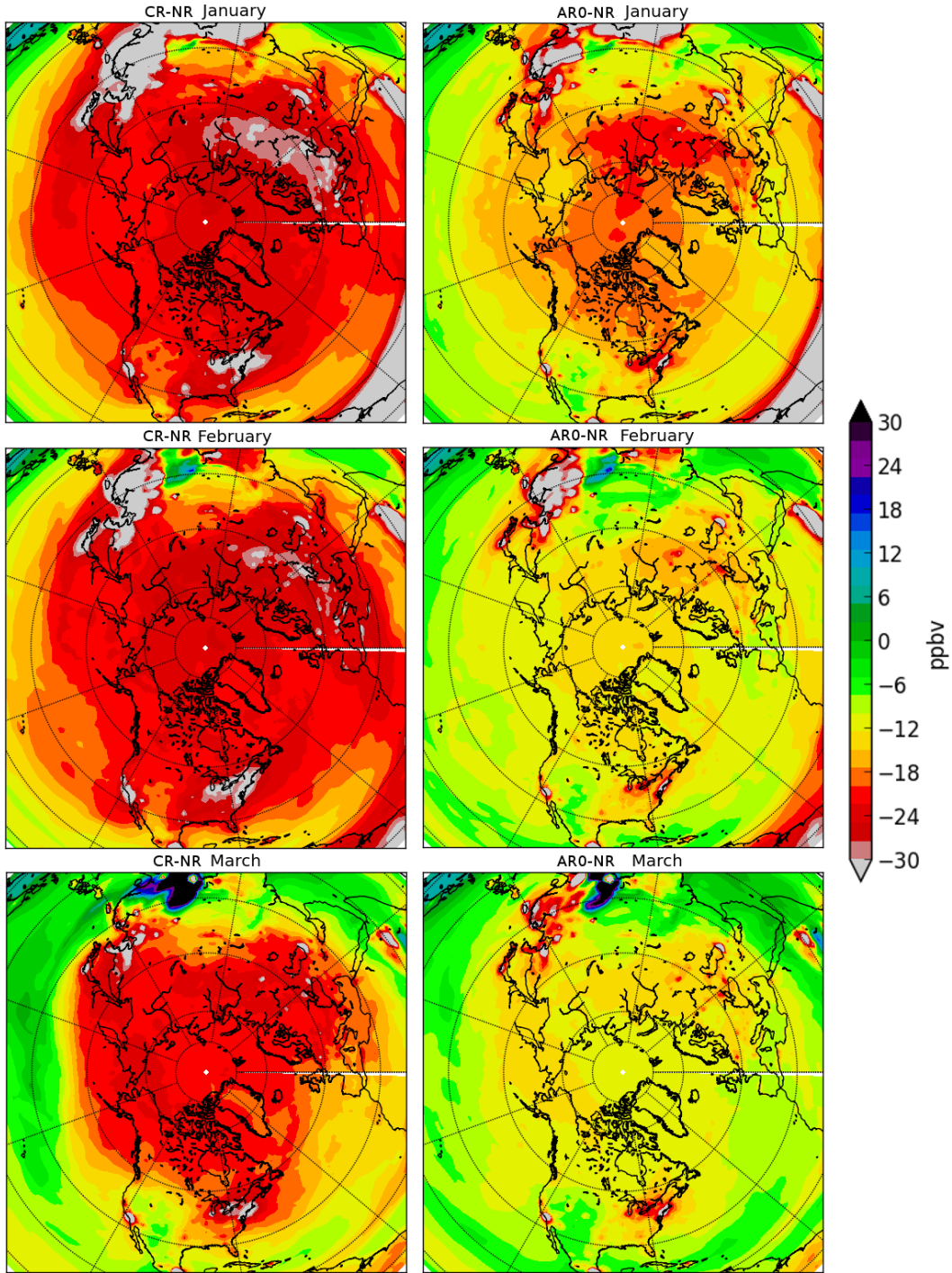




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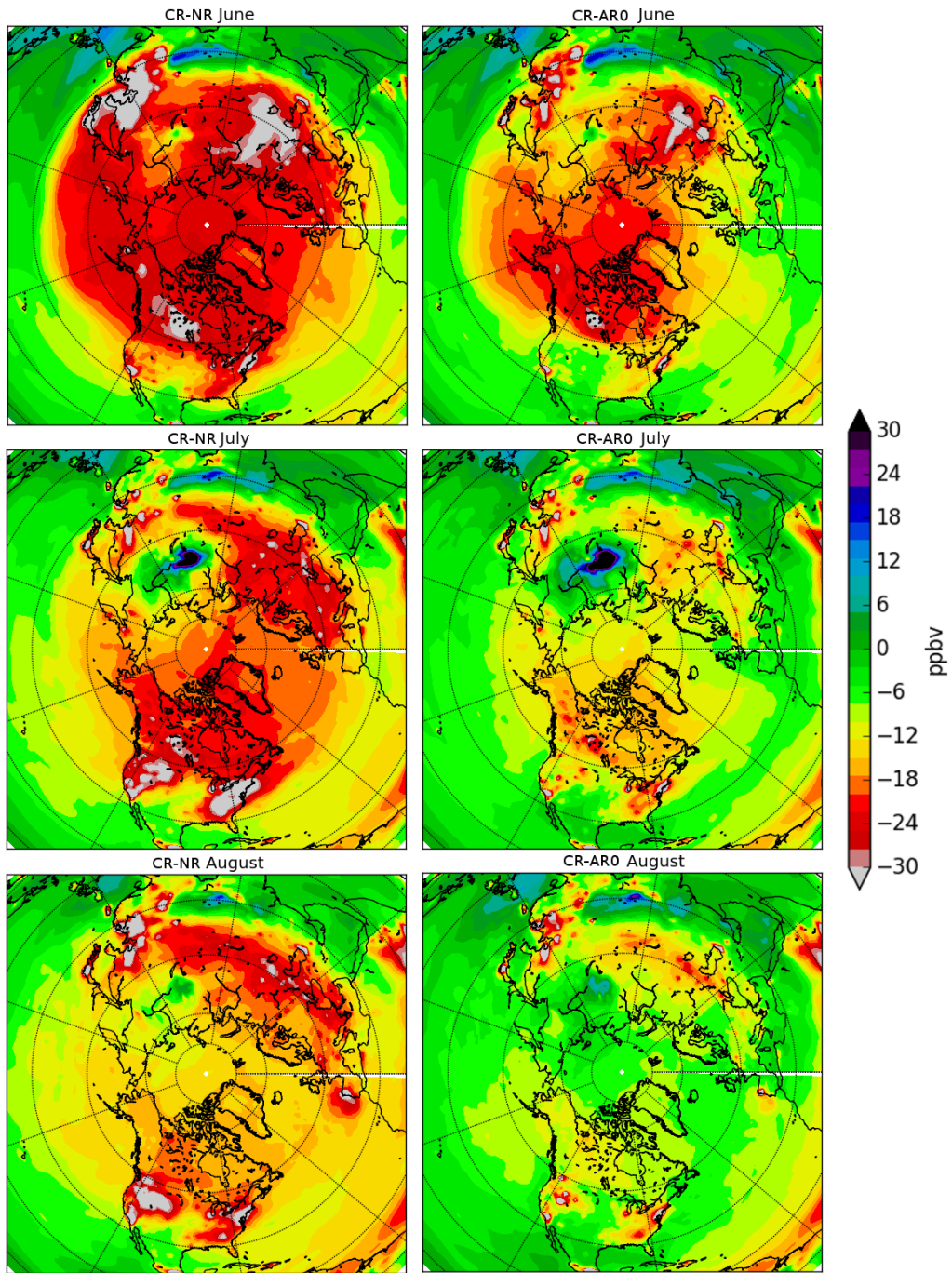
**Figure 4. RMS of relative increments in % (posterior state minus prior state divided by the prior state) between the surface to 200 hPa during January, February and March 2006 (top to bottom, left) and during June, July, August 2006 (top to bottom, right). Red and blue colors refer to relatively high and low values, respectively.**

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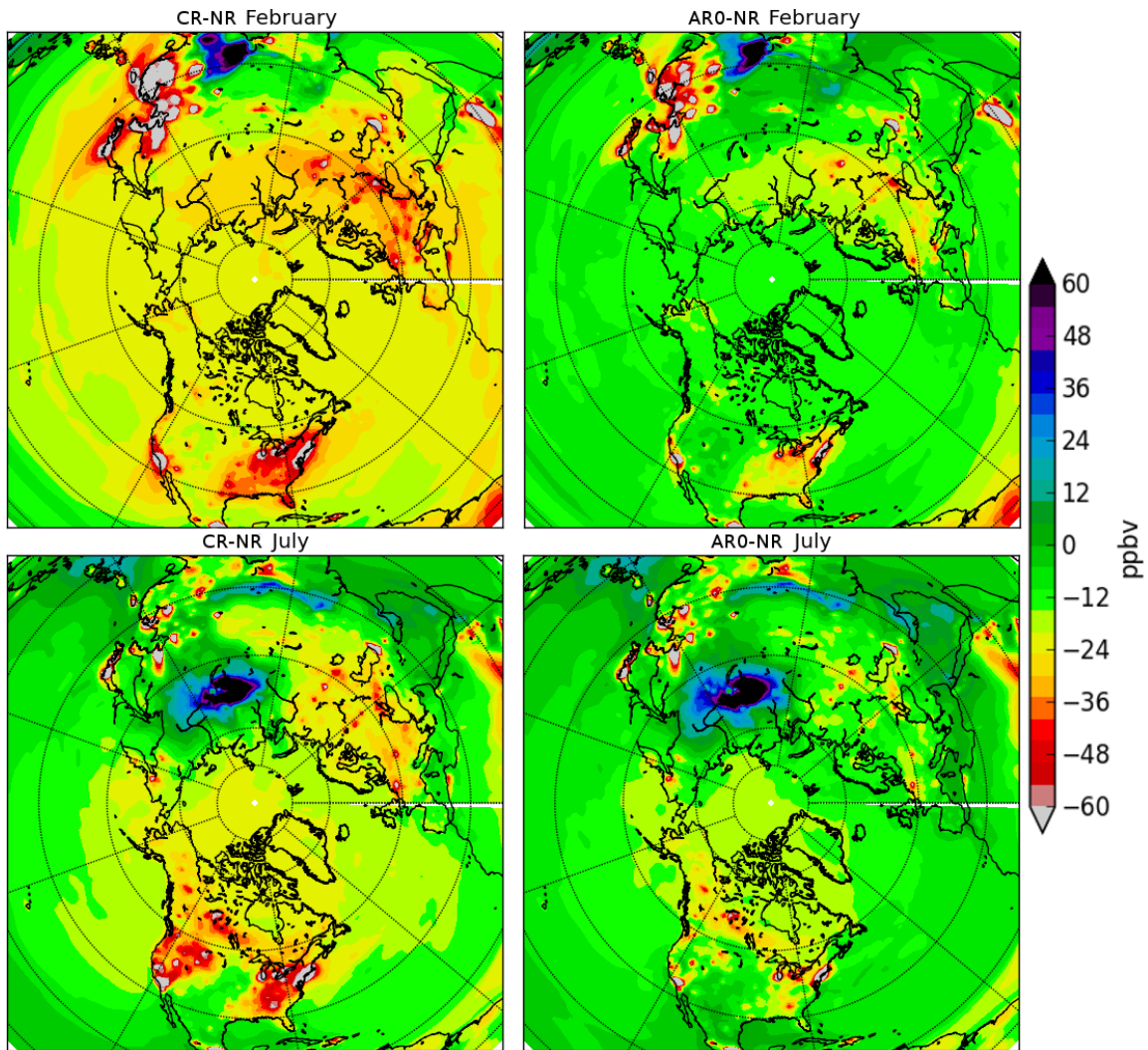
**Figure 5. Average differences in the tropospheric (surface to 200 hPa) CO fields between the control run (CR) and the nature run (NR) on the left hand side and average differences between full constellation assimilated CO (ARO) and the nature run (NR) on the right hand side during January, February and March (top to bottom) 2006. Units are ppbv.**



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**Figure 6. Same as Fig. 5 but for June, July and August 2006.**

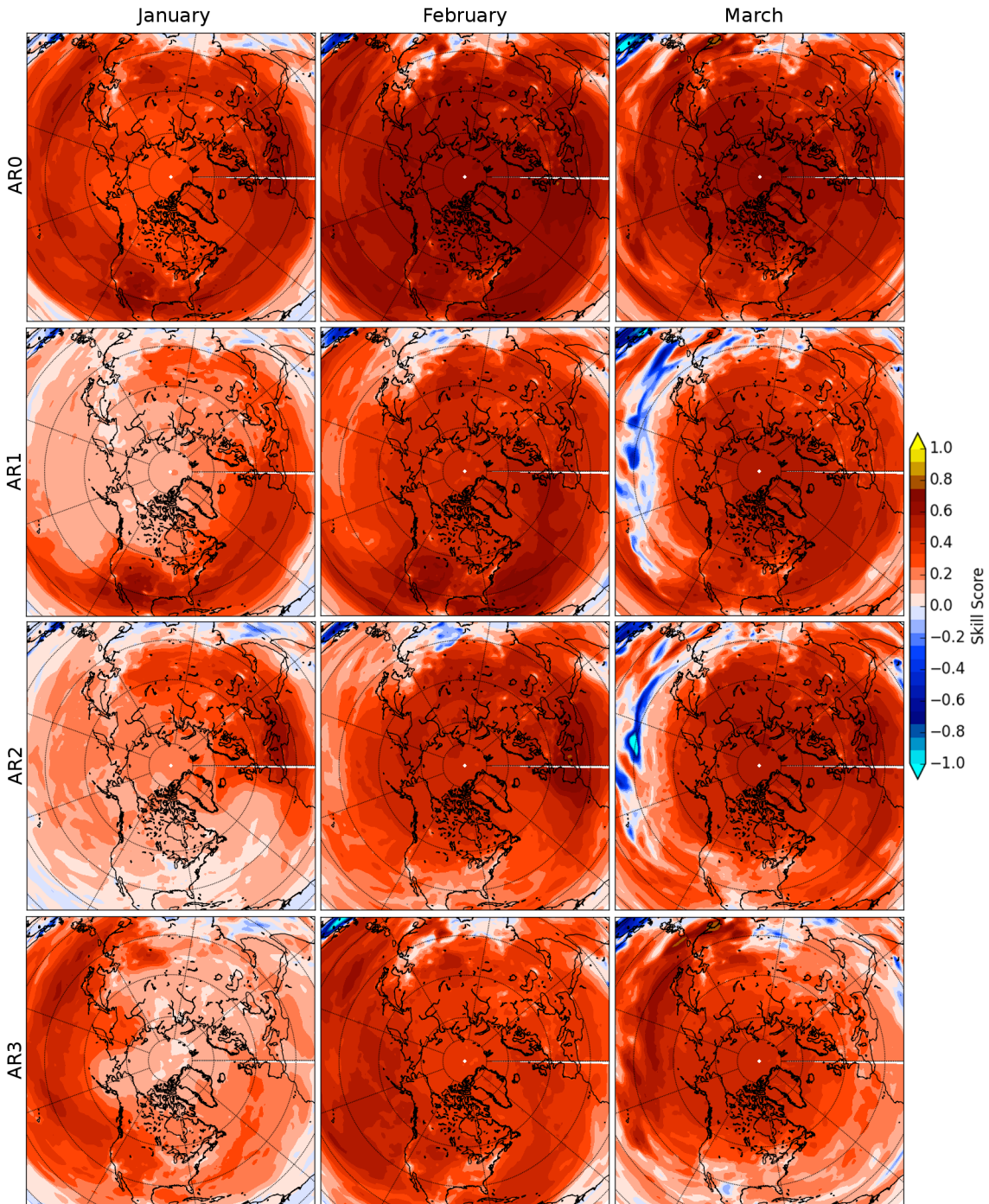




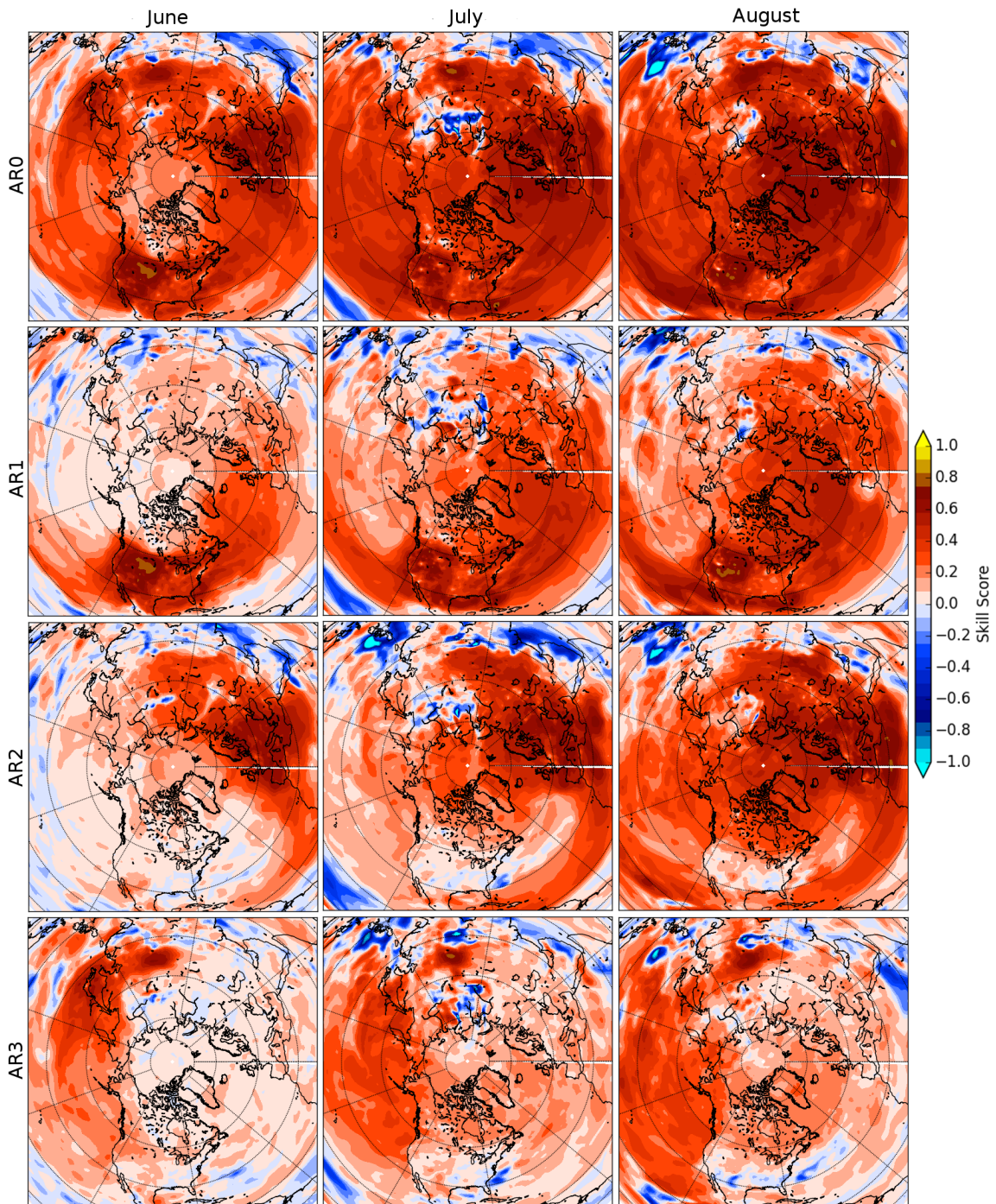
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**Figure 7. Average differences in the lower tropospheric (surface to 800 hPa) CO fields between the control run (CR) and the nature run (NR) on the left hand side and average differences between full constellation assimilated CO (ARO) and the nature run (NR) on the right hand side during February and July (top and bottom, respectively) 2006. Units are ppbv.**





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 812 **Figure 8. Assimilation skill scores (see text for details) for the full**  
 813 **constellation assimilation (AR0, first row), GEO-US assimilated only (AR1,**  
 814 **second row), GEO-EU assimilated only (AR2, third row) and GEO-AS**  
 815 **assimilated only (AR3, fourth row). Surface to 200hPa and monthly statistics**  
 816 **are performed during winter: January (first column), February (second**  
 817 **column) and March (third column) 2006. Red and blue colors refer to positive**  
 818 **and negative skill scores, respectively.**  
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**Figure 9. Same as Figure 8. but for summer: June (first column), July (second column) and August (third column) 2006.**