Response to Comments of Referee#1

Dear Reviewer:

We would like to thank you for the valuable and constructive comments/suggestions which helped to improve our manuscript. We have carefully revised the manuscript accordingly. Please find our point-to-point responses below (line numbers and figure numbers refer to the new version of manuscript; reviewer comments and suggestions are in italics, responses are in plain font; revised sections in the manuscript text in response to the comments are marked with red color).

1. In Section 3, was the WRF simulation nudged towards FNL analysis data? If so, is it fair to compare nudged model results with observations? Please clarify on this.

Response: We thank the reviewer for pointing out this missing information. In this study, a nudging towards FNL analysis data was used for the domain1 (75 km) and domain2 (15 km) to provide a more accurate meteorological boundary for domain3 (3 km). The innermost domain was driven one-way by initial and boundary inputs from the outer domain. No nudging was used in the domain3 (3 km). To evaluate the impact of aerosol-induced perturbations, we used only the meteorological fields in domain 3.

We have added clarifications on this point (Page 6, line 156): 'No nudging was used in the innermost domain. The aerosol-induced perturbations were estimated with the meteorological fields simulated in domain3.'

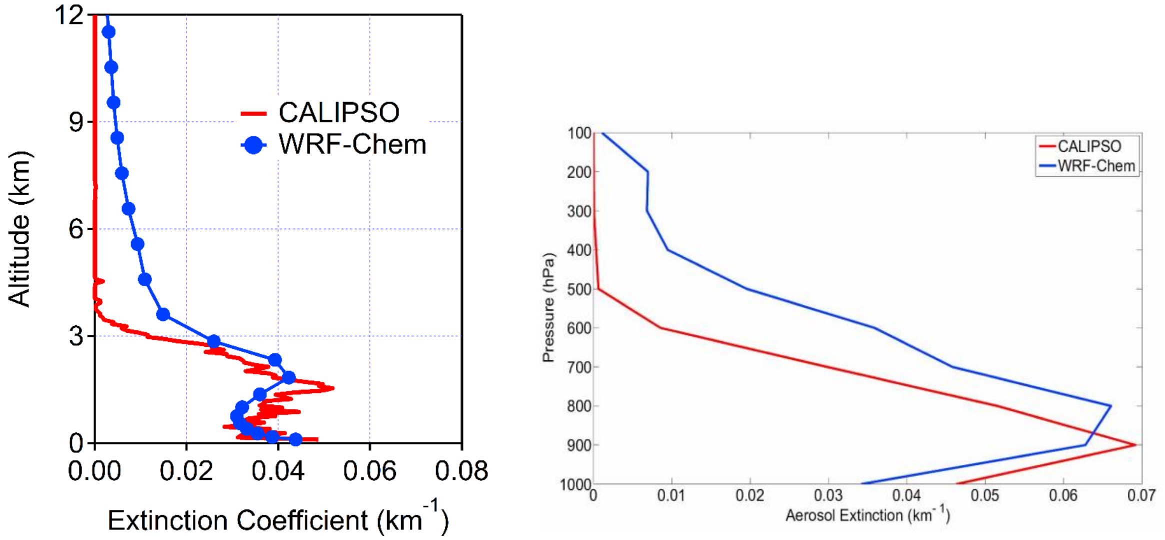
2. Is the conclusion sensitive to different plume rise parameterizations? Can the authors provide some validations on the simulated plume rise heights?

Response: Thanks for drawing attention to this point. The plume rise height is indeed an important factor for investigating aerosol-radiation-cloud interaction. For example, Johnson et al. (2004) showed the vertical distribution of light-absorbing aerosols could significantly affect the aerosol radiative effect. The plume rise parameterization used in this study is the only option provided in WRF-Chem (Grell et al., 2011; Freitas et al., 2007), which has shown commendable performance in simulating the plume height in the Amazon (Wu et al., 2011). To demonstrate the performance of the plume rise scheme in our study, we have conducted a comparison of the aerosol vertical distribution using CALIPSO observations.

The clear-sky monthly mean aerosol extinction profiles at 532 nm, provided by the CALIPSO Level 3 aerosol product (Tackett et al., 2018), was used here. The simulation data was processed to align with the observation by using outputs corresponding to the passing time of the satellite, excluding cloudy grid cells with a cloud criterion of 1 g/kg and interpolating the extinction coefficient at 550 nm to 532 nm. The averaged aerosol extinction profiles from the model results of domain 3 and observation were then compared (Fig. R1a).

The observed vertical profile of the aerosol extinction coefficient is basically reproduced by the model. The model well captures the peaks of the aerosol extinction coefficient at the surface and near 2 km. Similar to our study, an overestimation of the aerosol extinction coefficient above 3 km was also simulated by Wu et al. (2011, Fig. R1b). The phenomenon found in both studies may be caused by an overestimated exchange between PBL and the free atmosphere by turbulent mixing and convective transport, an underestimation of precipitation scavenging, and/or an overestimated plume rise at some fire spots. Generally, the vertical distribution of aerosols is captured reasonably by the model, which illustrates an acceptable performance of the plume rise parameterization and indicates the reliability of the conclusions about aerosol-radiation-cloud interactions obtained by the model.

We have added the comparison of the aerosol extinction coefficient profile to the aerosol evaluation section in the SI (Page 6, Line 142).



(a) (b)

Figure R1. (a) Monthly mean clear-sky aerosol extinction coefficient at 532 nm averaged over domain3. (b) Clear-sky aerosol extinction coefficient at 532 nm averaged over the Amazon Basin (Figure 3 from Wu et al. 2011).

3. Is the conclusion sensitive to the choice of the period? One month seems to be quite short. Why not consider multi-month or multi-year analysis?

Response: We acknowledge the reviewer's concern and we agree that longer term simulation (multi-month and/or multi-year) may add to the robustness of the conclusions. Currently, the simulations for this study were conducted for one month but not for a longer time mainly because of the limitation of the computing resources. As the WRF-Chem model itself is computation-demanding due to many coupled modules, and since additionally the simulations included 3-nested domains with fine resolution (3 km) at the innermost domain of 161*161 grids, and a set of 10 parallel sensitivity cases for 5 emission scenarios, the simulations are highly computational intensive.

Previous WRF-Chem simulations using such fine resolution to estimate the aerosol-radiation-cloud interaction were limited to 3-day to 8-day periods (Archer-Nicholls et al., 2016; Wu et al., 2011). The authors made an attempt in this study to expand the simulation time in order to include more cloud and precipitation cases and to give more robust constraints on the aerosol effect assessments compared to previous simulations. Limited by current computing resources, we chose a simulation period of 1 month, which has been shown to be a timescale that has short-term climatic significance (Becker et al., 2013).

The one-month simulation period targets September 2014 in order to make sure the study is conducted under typical dry season conditions. Pöhlker et al. (2016) comprehensively compared the meteorology over 18 years (1998-2016) and found that 2014 is a typical year and September is a typical month of the dry season for the Amazon area, e.g., the precipitation rates on September 2014 were found comparable to the 18-year average data and showed no pronounced hydrological anomalies (Fig. R2). Hence, this sensitivity study based on September 2014 can serve to reveal the typical sensitivity behavior of the dry season radiation, cloud, and precipitation over the central Amazon to BB aerosols. To address the questions of the representativeness of the simulation period and sensitivity of the results to other dry season periods, we added a statement on Line 91 and included a longer period simulation as a suggestion for future investigation on Line 460, respectively:

‘Comparison of the precipitation in central Amazonia in the year 2014 with that averaged over 18 years (1998–2016) indicates that the atmospheric conditions in this region in 2014 are climatically representative (Pöhlker et al., 2016). Therefore, the present study based on September 2014 may serve to represent the typical sensitivity behavior of the dry season climate to BB aerosol concentration variations.’

‘Furthermore, the sensitivity of the climate response to the concentration of BB aerosols may be influenced by the meteorological conditions, and as this study is based on September 2014, continuing model investigations based on varying and longer periods are needed to characterize the influence of variations in meteorology and to provide climatic assessments.’

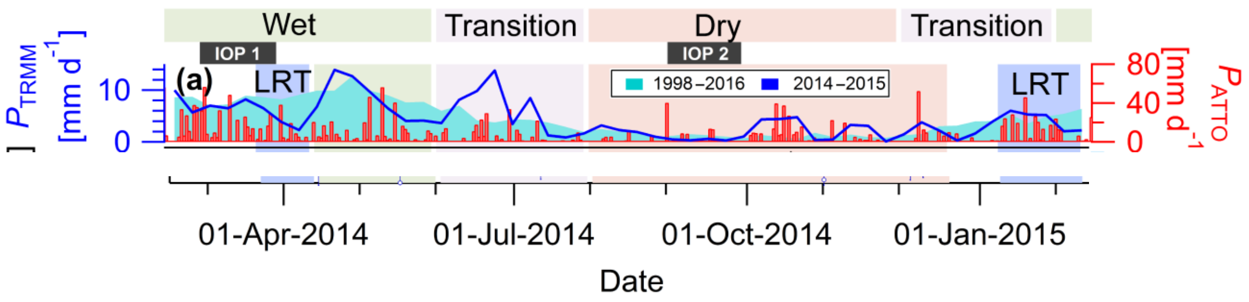


Figure R2. Precipitation rates from tropical rainfall measuring mission (TRMM) PTRMM and in situ measurements at the ATTO site PATTO. The P_{TRMM} seasonal cycles are derived from an area upwind of the ATTO site (59.5 W, 2.4 N, 54.0 W, 3.5 S), covering a long-term period from 1 January 1998 to 30 June 2016 (aqua shading), and the period of the CCN measurements from 1 March 2014 to 28 February 2015 (blue line). (Figure 1 from Pöhlker et al., 2016)

4. The main conclusion is that lower precipitation is expected with biomass burning aerosols. Do historical observations support this conclusion?

Response: The authors appreciate this insightful comment. Practically, the inconsistent methodologies used in observational data analysis and model simulation make it difficult to reconcile observation and model results. The modelling study uses strict control experiments by increasing/decreasing aerosol concentration on a fixed meteorology field. In contrast, satellite and in-situ observations usually use a method of sampling spatial and/or temporal correlation between aerosol and precipitation, and therefore other factors such as concurrent meteorological influence may bring in uncertainties. Besides, mismatched domain, study period, and convection stage between observation and model, e.g., that satellites usually measure at a specific time of day, should also be considered. Observational studies on the impact of biomass burning aerosols on precipitation, especially in-situ observations, are still scarce. The authors did find satellite observations in Rosenfeld et al. (1999) and Koren et al. (2012) showing overall consistent results with the conclusions in this study. Satellite measurements of a biomass burning episode in the tropical area by Rosenfeld et al. (1999) found that the rain formation process was shut down by biomass burning aerosols. This is consistent with our model results that biomass burning aerosols cause decreased precipitation occurrence and consequently lower precipitation amount. Another satellite measurement over the Amazon from June to August 2007 by Koren et al. (2012) showed a trend of decreasing precipitation due to biomass burning aerosols at an aerosol loading similar to this study (AOD >0.2). Yet, due to uncertainties caused by uncoordinated observation measurement and model simulation, the authors would be cautious and conservative about directly comparing the observations with the modelling results. More observation measurements and observation conducted in coordination with modelling might still be needed to provide constraints on aerosol-precipitation interactions in the model.

5. Why did an underestimation of precipitation during Sept 17 & 18 lead to much lower temperature and higher RH, compared to the observations? Why did the temperature vary little these days? Maybe it would be worthy to check the synoptic pattern and assure that this is not a model bug.

Response: Compared to the observations, the model underestimated the precipitation at the ATTO site during Sept 17 & 18 (Fig. S3), and meanwhile the temperature (Fig. S2a) in the model (red line) is higher than the observations (dot) and the RH (Fig. S2b) in the model (red line) is lower than the observations (dot). This may be associated with the fact that the precipitation itself moistens the soil, enhances the latent heat flux, and therefore leaves less net energy in the ground to heat the surface air, so low surface air temperature occurs during precipitation. Hence, the underestimation of precipitation in the model was accompanied by higher modelled temperature. As the RH is inversely related to air temperature, a lower modelled RH was found when precipitation was underestimated. We have added more explanation about the relationship between the bias in precipitation and in temperature and RH (Line 78 in SI): ‘This is expected since precipitation enhances surface evaporation and latent heat flux, leaves less net energy in the ground to heat the surface air (Zhuang et al., 2017), and therefore corresponds to a cool and moistened near-surface atmospheric state as can be found in the ATTO observation (Fig. S2). Consequently, the precipitation underestimation in the model was accompanied by higher biased simulated temperature and lower biased RH.’

The discrepancy of precipitation, temperature, and RH on Sept 17 & 18 between model and observation is obvious at the ATTO site but is not evident for the domain averaged precipitation (Fig. S6, comparison between simulated domain precipitation and TRMM observation). As suggested by the reviewer, a check of the synoptic pattern against the NCEP reanalysis was conducted (Fig. R3, shown below) and it shows the model basically captured the synoptic patterns. This means the model simulated the synoptic patterns and regional precipitation well but underestimated the precipitation at the ATTO site. The reason for this precipitation underestimation at the ATTO site could be associated with a location bias of rainfall. We then extracted the air temperature and RH from the grid (north of ATTO) where significant precipitation occurred on Sept 17&18 and compared it with the ATTO observations (Fig. R4, shown below). The air temperature (RH) at the precipitating grid dropped (increased) comparably to the observation during the rainfall period. It also serves as an indicator that the location bias of the rainfall on Sept 17 & 18 could account for the discrepancy of the meteorological factors between the simulation and the ATTO observation. We have added more explanation for the underestimation of precipitation at ATTO (Line 113 in SI): ‘The regional rainfall events on 6, 8, 17 and 18 Sep are well predicted by the model with a slight underestimation, which reflects a better model performance compared with the evident model underestimation of precipitation at the ATTO site on these four days. Moreover, the modelled synoptic patterns corresponding to the precipitation episodes are consistent with the NCEP reanalysis data (not shown). The well-reproduced regional synoptic and precipitation conditions in the model serve to corroborate that the precipitation underestimation at the ATTO site is likely induced by a local bias of rainfall location and neglecting precipitation of sub-grid convection by the model.’

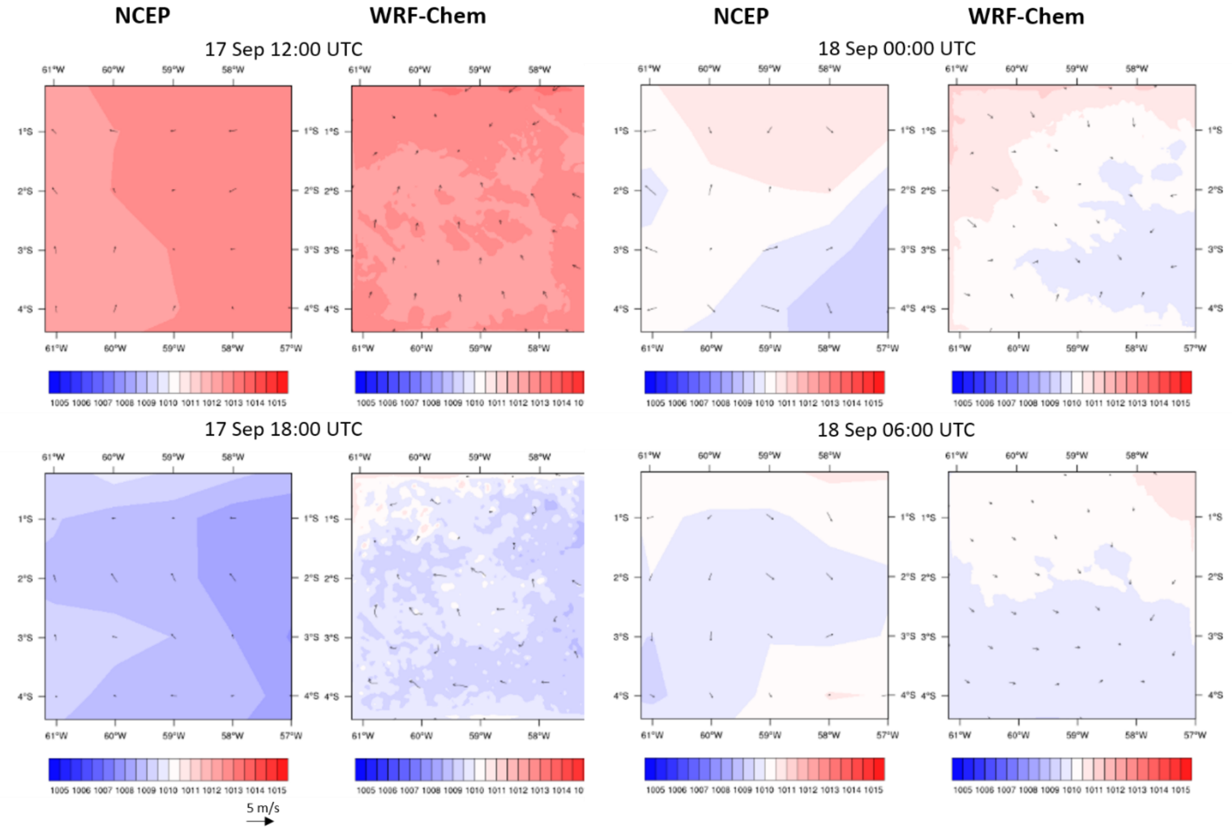


Figure R3. Surface air pressure and horizontal wind from NCEP reanalysis and WRF-Chem model.

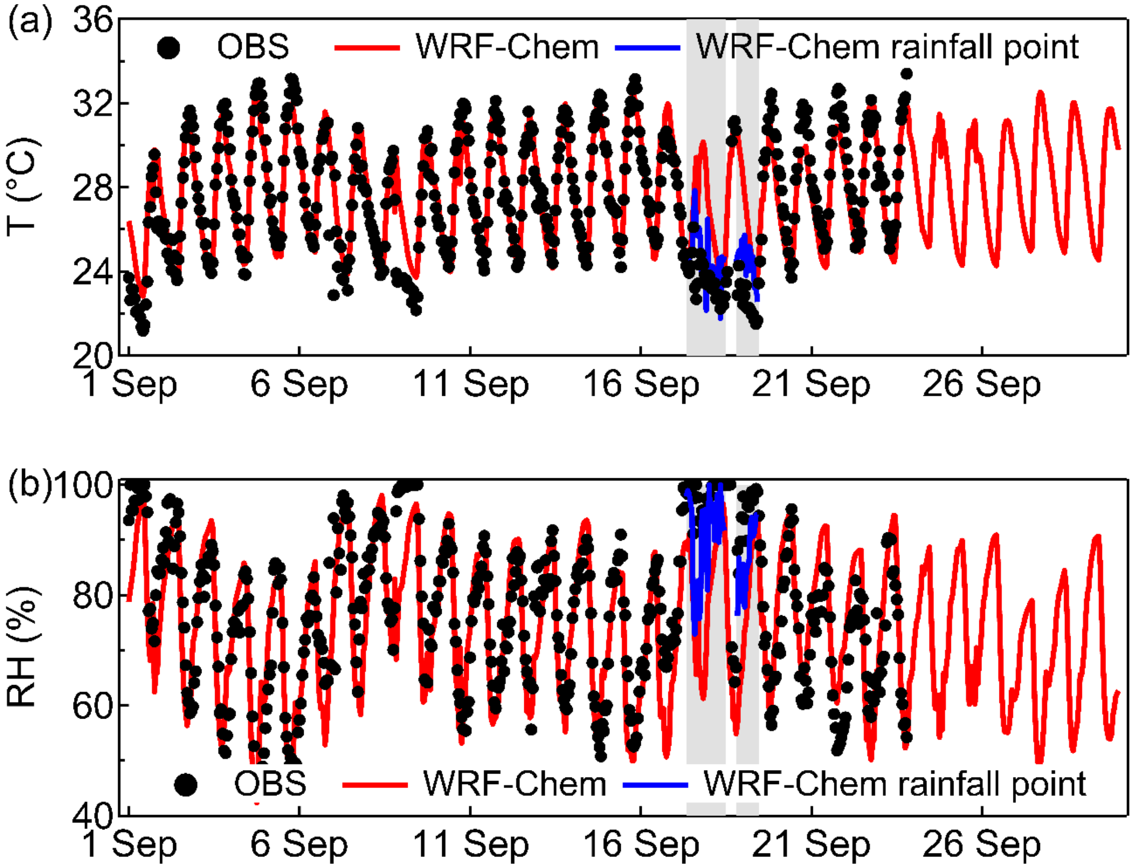


Figure R4. Time series of surface air temperature (a), relative humidity (b) from ATTO observation (black dot) and the domain3 simulation at ATTO (red line) and at significant precipitaiton point (blue line) during September 2014.

6. The validations of AOD simulation are not so impressive, it would be helpful to show: 1. the mean absolute bias & correlation between simulation and observation, making the validation more quantitative; 2. Reference other papers for the bias between simulated and observed AOD in South America and other regions, quantitatively.

<u>Response:</u> Thanks for the suggestion. We have revised the AOD evaluation in the studied domain according to the reviewer's suggestion to make it more clear and quantitative (Line 123 in SI).

'Table S3 shows the comparison of the modelled AOD against the AERONET observation at Manaus_EMBRAPA, a forest reservation site representative of the central Amazon environment (Artaxo et al., 2013). The model simulation generally captures the absolute value and the temporal variation of the observed AOD, with the mean bias and correlation coefficient being -0.03 and 0.54, respectively (Table S3). This is basically consistent with the AOD prediction accuracy in the Amazon by global models using the same fire emission inventory (Reddington et al., 2019; Pan et al., 2020). The slightly low bias in the AOD value could be related to an underestimated

> BB emission intensity due to errors in the detection of fires by satellite (Rosario et al., 2013) and/or an underestimation of the transatlantic transport from Africa (Holanda et al., 2020). Besides, the lack of SOA production in the model may also account for the bias in the AOD simulation (Bond and Bergstrom, 2006).’

Table S3. Comparison of AOD and SSA at 550 nm obtained from model simulation in domain3 and observation.

	Observation	Model[a]
	AOD	
Manaus_EMBRAPA (AERONET)	0.24±0.10 (average of Sep 2014)	0.21±0.05 (R[b]=0.54)
	SSA	
TT34[c] (Rizzo et al., 2013)	0.87±0.06 (average of Jul–Dec 2008–2010)	0.89±0.01
ATTO[d] (Saturno et al., 2018b)	0.88 (average of Aug–Nov 2012–2017)	0.90±0.01

a) Model results with EMIS1, averaged for September 2014.
b) R represents the correlation coefficient between the observation and model simulation.
c) The SSA values at this site are for 637 nm. Calculation of SSA at 550 nm is not conducted due to incomplete information on Angstrom exponent in Rizzo et al. (2013).
d) The SSA observation for the ATTO site is obtained from Saturno et al. (2018b) by extrapolating the original value at 637 nm to that at 550 nm using the Angstrom exponents in Saturno et al. (2018b).

7. The authors may consider to move Section 3 to supplement to make the manuscript less length and more focused.

Response: Very good suggestion. We have moved Section 3 to SI and added a short summary of the evaluation before the results section (Line 208).

‘The WRF-Chem simulation with the EMIS1 scenario was evaluated for the meteorological conditions and the aerosol field using ground-based, radiosonde, and satellite remote sensing measurements (see Supplement Text S1–S3). The results show that the model simulation at 3 km resolution reasonably reproduces the metrological field in terms of surface conditions, vertical atmospheric structure, and regional precipitation. The total cloud fraction and liquid cloud amount are well captured by the model while the simulated ice water amount shows lower magnitude than the observations. The model generates close agreement of the predicted aerosol properties with the observations, including the aerosol optical properties (AOD and SSA) and the CCN concentrations at different supersaturation conditions. Details of the model evaluation are provided in the Supplement. The satisfactory performance of the model enables it to provide reliable assessments of the BB aerosol effects on the regional climate through aerosol-radiation-cloud interactions.’

8. Technical comments: Line 27: ’which enables them’ -> ’which enable them’

Response: Accepted.

9. Technical comments: Line 151: Need a reference

Response: Accepted. The reference has been added at Line 158 of the revised manuscript:

> 'Anthropogenic emissions were from the EDGAR-HTAPv2, a global gridded air pollution emission dataset with a resolution of 0.1° × 0.1° (http://edgar.jrc.ec.europa.eu/htap_v2; Janssens-Maenhout et al., 2015).'
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> Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, Atmos. Chem. Phys., 15, 11411–11432, https://doi.org/10.5194/acp-15-11411-2015, 2015.

Reference

Becker, E. J., H. van den Dool, and M. Peña, 2013: Short-Term Climate Extremes: Prediction Skill and Predictability. J. Climate, 26, 512–531, https://doi.org/10.1175/JCLI-D-12-00177.1.