Anonymous Referee #1

Zhao et al. present a study on total column CO_2 and CH_4 in Berlin. Their results from a high-resolution modelling framework based on WRF-GHG-GFS and VPRM-EDGAR fluxes is compared to previously published observations by Hase et al.2014. The authors found that XCO_2 can be modelled reasonably well, while CH_4 showed a significant bias of ca. 2.7%. Using a differential column methodology the influence of variations in the boundary conditions and non-anthropogenic sources can be reduced and the model-data mismatch of the DCM-derived XCO_2 and XCH_4 was further investigated.

We thank the anonymous Referee #1 for their time and valuable comments to improve this manuscript. The general and specific comments are addressed by point-by-point detailed replies below. Referee's comments have been repeated in black. Author's replies and edited contents are marked in blue and red, respectively. We would like to add Frank Hase and Matthias Frey as the co-authors in this paper because of their contributions in the measurements and in clarifying our analysis during the review phase.

General comments

Overall, the paper is well-written and the structure is straightforward. Despite only covering a few days of measurements this paper adds interesting results to a growing field of research and demonstrates the value of DCM. After addressing the general and specific comments, I would recommend this paper for publication.

We thank the anonymous referee #1 for the careful reading of the manuscript and helpful comments. We have addressed the specific and general comments.

The manuscript falls a bit short by not considering or discussing all potential sinks and sources of CH_4, but their assumptions are largely supported by citations and other studies. Given the high temporal and spatial resolution of the modelling framework it would have also allowed to investigate other issues in more detail, e.g. which sources in the region are dominant (are the power plants the key contributors to CO_2,ff in Berlin) or how would a change in the daily emission cycle affect the model-observation mismatch. The spatial and temporal dis-aggregation is mentioned as a major source of uncertainty in bottom-up inventories for cities, but this topic is not really discussed through the lens of the modelling and measurement results here.

In this study, we do not cover all the processes related to the tracers and mainly focus on the dominant emission tracers for CO_2 and CH_4 within urban areas. In the 'online' tracer calculation for anthropogenic emissions, the CO_2 and CH_4 surface fluxes are derived from the EDGAR V4.1 emission inventory, and mixed vertically and horizontally based on the meteorological field. We make the assumption that the attributions from EDGAR V4.1 are reliable in our study. In view of the proportions of different emission sectors in EDGAR V4.1, energy industry and road transportation are the key contributors to CO_2 anthropogenic emissions in Berlin while extraction and distribution of fossil fuels are the largest CH_4 emitters. For the spatial and temporal dis-aggregation, the coarser emission inventory could ignore the key emission points potentially (Line 139-149), and the meteorological data might not be able to capture the entire transport features of air masses (Line 182-185).

Specific comments

Line 23: 50% seems to be an extreme example and it seems advisable to give the range and typical uncertainty of national emission inventory reports.

Response: We thank the referee for pointint it out. Indeed, 50% is a specific value. The range of the uncertainty on individual regional and national total fossil-fuel carbon dioxide emissions is from a few percent to more than 50% (Andres et al., 2012). We have re-phrased the sentence as follows (Line 32-34):

This approach has some uncertainty, e.g., on the national fossil-fuel CO_2 emission estimates, ranging from a few percent (e.g., 3%-5% for the US) to a maximum of over 50% for countries with less resources for data collection and poor statistical framework (Andres et al., 2012).

Line 37: Typical ‘top-down’ methods rely on prior information on fluxes, therefore, the assumption that they are ‘independent’ should be further explained.

Response: Thanks for pointing out this unclear description. Estimates from the ‘top-down’ approach on local to global scales do rely on bottom-up estimates as priors. Advanced measurement technologies and high-resolution models are able to estimate regional emissions on the basic of the ’top-down’ approach such that regional or national bottom-up emission inventories can be assessed and verified (Wunch et al., 2009; Montzka et al., 2011; Bergamaschi et al., 2018). Thus, the ’top-down’ approach can help to identify the discrepancy compared to the ’bottom-up’ approach and highlight the uncertainties in both methods. We have re-phrased it in Lines 37-38 as follows:

The ‘top-down’ approach can not only provide estimated global fluxes, but also verify the consistency and assess the uncertainties of bottom-up emission inventories (Wunch et al., 2009; Montzka et al., 2011; Bergamaschi et al., 2018).

Line 43: Please clarify what kind of ‘carbon cycle processes’ you are referring here.

Response: The phrase ’Carbon cycle process’ describes the process in which carbon, released from emission sources, travels from the atmosphere to organisms and the earth and then back into the atmosphere, i.e., carbon sources and sinks. We have clarified in Line 42-45:

Such measurements, i.e. measurements of concentration averaged over a column of air, are performed to help to disentangle the effects of atmospheric mixing from the surface exchange (Wunch et al., 2011) and decrease the biases associated with estimates of carbon sources and sinks in atmospheric inversions (Olsen and Randerson, 2004).

Line 53: Please add information about the manufacturer for the EM27/SUN

Response: We have included the manufacturer for the EM27/SUN in Line 53-54:

Chen et al.(2016) applied the DCM using compact Fourier Transform Spectrometers (FTS) EM27/SUN (Bruker Optik, Germany).

Line 61: Vogel et al. 2018 also seem be using a very similar upwind versus downwind approach.

Response: Vogel et al. (2018) applied a similar upwind-downwind approach to determine the major contribution of CO_2 in urban area using the case of Paris. In their station-to-station calculations, one site (RES) is taken as the upwind reference to assess the impact of local sources (details in Section 3.2.3 of Vogel’s paper), whereas we considered the wind direction when selecting the downwind and upwind sites in our case. The details are discussed in the conclusion (Line 374-390) of the content.

Line 68: Previous studies have found that urban carbon fluxes are significantly higher than predicted with conventional models like VPRM (Hardiman et al.2017: https://doi.org/10.1016/j.scitotenv.2017.03.028). Using VPRM is thus a limitation that should be discussed.

Response: Thank you for your suggestions. We discussed this line in Lines 72-75 as follows:

Biogenic carbon fluxes given by the VPRM model tend to underestimate urban ecosystem carbon exchange, owing to the incomplete understanding of urban vegetation, and to conditions related to urban heat islands and altered urban phenology (Hardiman et al., 2017).

Line 76: It would seem important to note that Berlin is actually a state, with more regulatory influence than other cities.

Response: Thanks for pointing it out. We have included the regulatory influence in Line 86-88, as follows:

With its strong regulatory influence as a ‘state’ within Germany, and a strongly supportive policy, Berlin has already transformed itself into a climate-friendly city in which CO_2 emissions have been reduced by a third compared with 1990 levels, aiming for carbon neutrality by 2050 (Homann, 2018).

Line 93: Please clarify what is meant by ‘actual meteorological conditions’.

Response: The actual meteorological conditions refers to a model initialization using real data for meteorological fields in the pre-processing, instead of idealized initialization. We have clarified in Line 104-106:

...based on the actual meteorological data in this case. The meteorological initial conditions and lateral boundary conditions were taken from the Global Forecast System (GFS) model reanalysis in which in-situ measurements and satellite observations have been assimilated.

Line 115 – eq. 1: The equation for CH_4 is missing any biogenic production of CH_4. Furthermore, why is the soil sink of CH_4 considered, but not the photochemical sink? Both are responsible for the CH_4 lifetime.

Line 227: The soil sink for CH_4 is significantly smaller the photo-chemical sink – why was soil uptake investigated here?

Line 198: Please clarify why marshy woodlands cannot be a source of biogenic CH_4?

Response: Thanks for these valuable comments about the CH_4 tracer analysis. We do agree that with a CH_4 lifetime of approx. 9 years, neither soil uptake nor photo-chemical sink would have a strong impact in our short simulation period (10 days), mentioned in Line 153-155 of the content.

The main reason why the soil-uptake process is taken into consideration in this study is that Berlin is located in an area of low-lying, marshy woodlands. We doubted that marshy woodlands soils might be potentially effective CH_4 sinks and can be quantified in the soil uptake model which was already built in WRF-GHG by Dr. Veronika Beck. As seen clearly in Fig.5 of the content, we concluded that the emissions from the soil-uptake process have almost no influence on the daily variations of XCH_4.

This soil uptake model built in WRF-GHG is a process-based model for the calculation of the consumption of atmospheric CH_4 by soils (the result of an entirely biological oxidation process, including the diffusion and microbial oxidation processes). Generally, through simplifying the physiology of the methanotrophs, this process-based model calculates the CH_4 fluxes into soil based on the activity of methanotrophs (i.e., the potential rate of CH_4 oxidation within the soil), a number environmental factors (soil temperature, soil type, moisture, etc..), the diffusivity of the topsoil, the first-order oxidation rate, etc.. The details can be found in Beck et al., 2011.

For the photo-chemistry in the troposphere, in turn, the chemical reaction of 'OH' production in the lower stratosphere and upper troposphere needs to be quantified. At the moment, the WRF-GHG model allows for passive tracer transport simulations, i.e. without any chemical reactions of CH_4 mentioned in Sect.2. In further studies we may consider to add an additional 'off-line' flux chemical model or the corresponding process-based model to quantify the changes in CH_4 concentrations from photochemistry and combine the estimates into our simulation, mentioned in the conclusion of the content.

Woodland soils can be definitely regarded as an effective CH_4 source. During the process of methanogenesis, methane is a byproduct in hypoxic conditions, which are common in wetlands, where they are responsible for marsh gas. We thank Dr. Michal Galkowski from Max Plank Institute for Biogeochemistry who provided us with the biogenic-related CH_4 emissions for the area closer to the city Berlin region (242 km × 202 km) based on simulations for 2018 using the updated version of WRF-GHG. In Dr.Galkowski's study, wetlands contributes around 15% of the total CH_4 emissions (including anthropogenic, dispersed sources and point sources, wetland and termites) for his domain. Wetlands sources in this domain are mainly attributed to the area in the southern part which is roughly near Biosphärenreservat Spreewald (approx. 60 km away from the Berlin center) and is not included in the innermost domain of our study. Meanwhile, as described in our content (Sect 3.4), there is no wetland in the city of Berlin according to the MODIS Land Cover Map. Thus, we can have an estimate about the magnitude of biogenic CH_4 emissions based on Dr.Galkowski's data and find that the influence for CH_4 biogenic emissions from wetlands is quite weak in our innermost domain. In this study, our interest is mainly on the major emission tracers. So we did not consider the wetland source as the targeted CH_4 emission tracer in the analysis for the city of Berlin.

Line 147: Were wind speeds in different heights also investigate? How could the Ekman spiral have affected your results?

Response: We did a brief analysis on the vertical distribution of wind fields. As seen from Fig.1, above approx.300 hPa, the lower the pressure is, the larger the wind speed is. While the wind speed decrease sharply with the increase of the

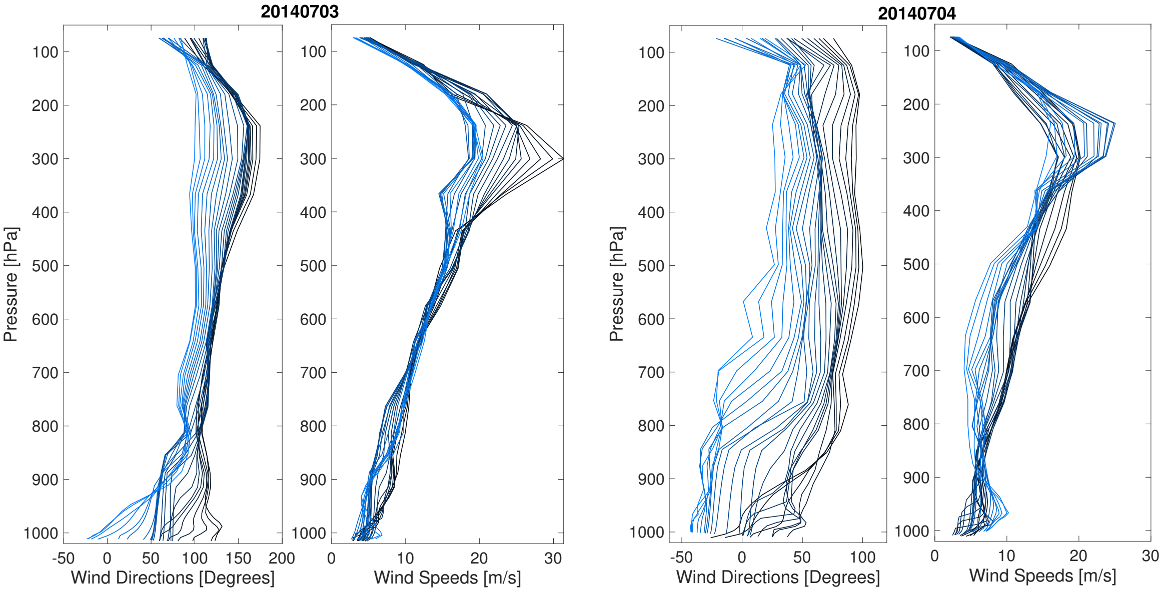

Figure 1: The vertical distribution of wind fields (wind speeds and wind directions) on 3rd July (left side) and 4th July (right side) in Tegel. The colors from black to blue represent the time from morning to evening.

height (below 300 hPa). Overall, wind directions shift from northeast to northwest from morning to evening. As depicted in the trend of the vertical distribution of wind directions, the surface wind shows a prevailing pattern towards the east with the increase of the altitude in the troposphere. Then above the troposphere, the prevailing pattern turns gradually to be in the opposite direction (the east). The vertical distributions of wind fields are also shown in the Appendix.E of the content.

Line 152+ : Why is only R^2 reported? It seems the root mean square difference would be an important measure of model performance here.
Line 268-269: Why is only R^2 given as metric of performance? Root mean squared differences could be important to investigate at all to properly judge the model-observation mismatch.

Response: Thanks for your valuable suggestions. We do agree with your point. Root mean square error (RMSE) is definitely a more appropriate measure to describe how accurately the model simulates and indicates the absolute value to show how simulated values are close to measure data points. Lower RMSE shows better fits generally. In our rephrased content, we have provided RMSEs in figures and RMSE also helps to evaluate the performance of two calculations in differential column method.

Line 159: ad → and

Response: Change made.

Line 210: What is meant by ‘background’ here? Do you consider the lowest point per day to be the background or does background refer to a theoretical Xgas value without any sources and sinks within Berlin (or a wider region)?

Response: Sorry for this unclear point. The ’background’ concentrations here are Xgas values without the influence from any sources within the domain area. Correspondingly, the total concentrations are the combination between the background concentrations and the concentration changes from different tracers. The background and total concentration fields are initialized by the CAMS dataset. We have clarified this point in Line 127.

Line 213: ‘owning’ → ‘owing’ – also ‘wiggles’ seems to be a fairly colloquial choice of words here.

Thanks for pointing it out. We have changed these two parts in Line 286.

Line 248: It seems the 10m wind speed is rather a ‘performance metric’ or ‘diagnostic parameter’ rather than a ‘standard’.

Response: We changed the standard condition for the selection of downwind and upwind sites into the simulated daily mean wind directions (see the left column of Fig.7). Details are discussed in Sect 4.1 and Table.1.

Line 251: What were wind conditions at higher levels of the PBL?

Response: As described in Sect.2, the vertical layers in WRF model follows the pressure definition and the upper pressure in our case is up to tropopause height. Figure.2 shows the wind fields within PBL on 3rd July. The PBL is situated either in the second or third layer (morning and night) or in the 13th layer (noon) in our domain. The wind speeds and wind directions closer to the PBL are generally higher than the surface wind fields.

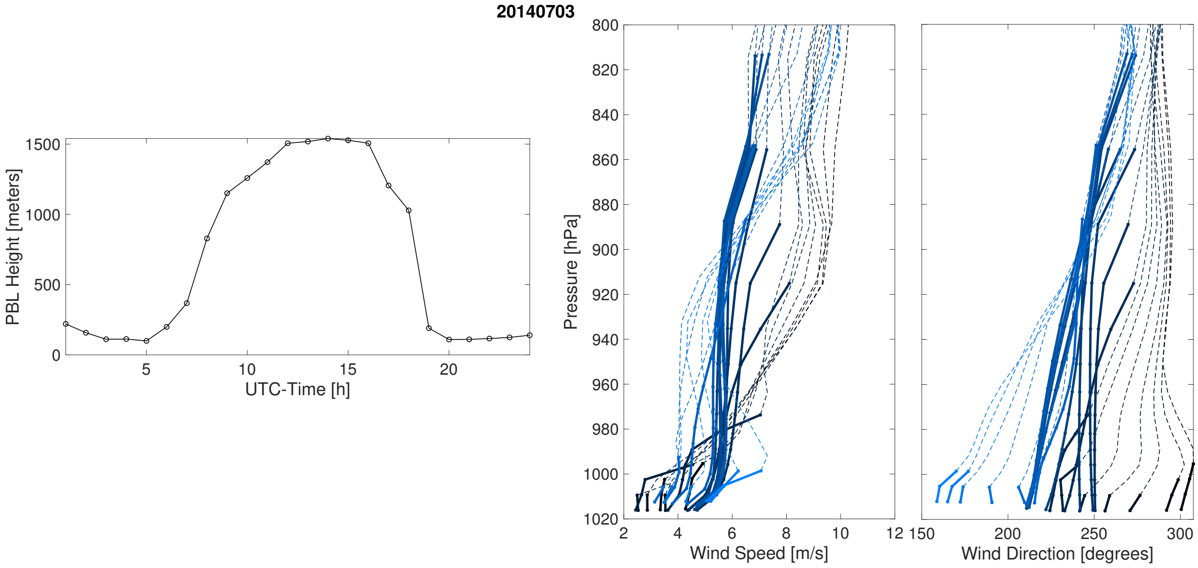

Figure 2: Variations of the simulated Planetary Boundary Layer (PBL) Height (left side), and the wind speeds (middle) and wind directions (right) within PBL on 3rd July at Tegel. The colors from black to blue represent the time from morning to evening. The bold solid lines represent the values within PBL.

Line 272: Why was this specific time window chosen?

Response: This time window was chosen because there is a wind switch at 10 meters in the measurements which was not seen in the simulations. In the rephrased content, we used the daily mean wind directions as the reference and re-define the calculation of delta concentrations. This daily mean wind directions average the hourly vertical-mean wind directions. This specific time window is not considered any more.

Line 295: Please clarify what is meant by ‘concentration fields’, I would assume the instrument measures one total column Xgas value for each time step and not a higher dimensional field.

Response: Thanks for pointing it out. The sentence has been re-phrased for ’The instruments measure the concentration value every minute (Hase et al., 2015).’

Line 310: Couldn’t this also be caused by errors in the spatio-temporal distribution of emission map or other just missing non-anthropogenic sources. Furthermore, the underlying (GFS-driven) wind data is also limited in its resolution in space and time.

Response: Thanks for your valuable suggestion. Indeed, these could be potentially the reasons why the simulations show stable variations compared to the measurements. The sentence in Line 250-253 has been rephrased:

The smaller variations from the simulation results can, e.g., be caused by the error from the spatial-temporal treatment of emission maps, underestimated emissions from anthropogenic activities, the coarse wind data and/or the smoothing of actual extreme values in the simulation.

Line 329: Please clarify the distinction you make between being ‘pivotal’ and playing a leading role.

Response: Sorry for this confusion. Basically, the words ’pivotal’ and ’leading’ both are used to emphasize the importance and the large contribution of one object. To avoid this confusion, we only keep ’pivotal’ in the content.

Line 331: Please clarify ‘background’. If this refers to a larger scale value without the influence of sources, then this is not surprising. You ignore photo-chemistry and assume only anthropogenic CH_4 sources.

Response: Thanks for pointing out this unclear point. Given the very limited size of the domain, ignoring the methane photo-chemistry would have at most a very small effect on this offset. As described above, the biogenic-related CH_4 emissions contribute very little to the CH_4 emissions, while the CH_4 bias is around 50 ppb. Furthermore, the background, as described above, is taken from CAMS, a global atmospheric composition analysis which takes the photochemical sink into account.

Line 345: What is a ‘flux framework’?

Response: The ’flux framework’ stands for a suite of models in the operational CAMS global assimilation and forecasting system, which is directly referred from Vogel et al., 2019.

Line 346: According to the CAMS webpage their CO_2 vegetation model is C-Tessel (https://atmosphere.copernicus.eu/global-production-system)

Response: Thanks for pointing it out and it has been corrected.

Figure 3: Symbols are hard to distinguish.

Response: Thanks for your suggestion and the figure has been re-plotted.

Figure 4: Consider changing label to ‘XCO2 enhancement from. . .’.

Response: The edits have been made in Fig.5.

Figure 6: East Wind > east wind; also please consider adding the administrative boundaries of Berlin or is all of the urban are in the plot Berlin?

Response: The edits have been made in the new figure.

Figure 10: The legend is hard to read and curve for the observations are hard to see as well. Also consider adding error bars to the observed values.

Response: The error bars have been added for all the measurement-related values for concentration fields and the legends are enlarged.

Line 460: → Why ‘assessed’?

Sorry for this wrong word. It should be ’accessed’ instead of ’assessed’. The ’accessed’ is used to show the time when the data or information was taken from the link.

References

Andres, R. J., Boden, T. A., Bréon, F.-M., Ciais, P., Davis, S., Erickson, D., Gregg, J. S., Jacobson, A., Marland, G., Miller, J., et al.: A synthesis of carbon dioxide emissions from fossil-fuel combustion, Biogeosciences, 9, 1845–1871, https://doi.org/10.5194/bg-9-1845-2012, 2012.

Bergamaschi, P., Karstens, U., Manning, A. J., Saunois, M., Tsuruta, A., Berchet, A., Vermeulen, A. T., Arnold, T., Janssens-Maenhout, G., Hammer, S., Levin, I., Schmidt, M., Ramonet, M., Lopez, M., Lavric, J., Aalto, T., Chen, H., Feist, D. G., Gerbig, C., Haszpra, L., Hermansen, O., Manca, G., Moncrieff, J., Meinhardt, F., Necki, J., Galkowski, M., O'Doherty, S., Paramonova, N., Scheeren, H. A., Steinbacher, M., and Dlugokencky, E.: Inverse modelling of European CH_4 emissions during 2006–2012 using different inverse models and reassessed atmospheric observations, Atmos. Chem. Phys., 18, 901-920, https://doi.org/10.5194/acp-18-901-2018, 2018.

Hardiman, B. S., Wang, J. A., Hutyra, L. R., Gately, C. K., Getson, J. M., and Friedl, M. A.: Accounting for urban biogenic fluxes in regional carbon budgets, Science of the Total Environment, 592, 366–372, https://doi.org/10.1016/j.scitotenv.2017.03.028, 2017.

Homann, G.: Climate Protection in Berlin, Tech. rep., Senate Department for the Environment, Transport and Climate Protection, https://www.berlin.de/senuvk/klimaschutz/politik/download/klimaschutzpolitik_en.pdf, 2018.

Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J.G., Dlugokencky, E.J., Bergamaschi, P., Bergmann, D., Blake, D.R., Bruhwiler, L. and Cameron-Smith, P., Three decades of global methane sources and sinks. Nature geoscience, 6(10), p.813, 2013.

Montzka, S. A., Dlugokencky, E. J., and Butler, J. H.: Non-CO_2 greenhouse gases and climate change, Nature, 476, 43–50, https://doi.org/10.1038/nature10322, 2011.

Olsen, S. C. and Randerson, J. T.: Differences between surface and column atmospheric CO_2 and implications for carbon cycle research, Journal of Geophysical Research: Atmospheres, 109, https://doi.org/10.1029/2003JD003968, 2004.

Ridgwell, A. J., Stewart, J. M. Keith, G.: Consumption of atmospheric methane by soils: A process-based model. Global Biogeochemical Cycles, 13.1, 59-70, https://doi.org/10.1029/1998GB900004, 1998.

Vogel, F. R., Frey, M., Staufer, J., Hase, F., Broquet, G., Xueref-Remy, I., Chevallier, F., Ciais, P., Sha, M. K., Chelin, P., Jeseck, P., Janssen, C., Té, Y., Groß, J., Blumenstock, T., Tu, Q., and Orphal, J.: XCO_2 in an emission hot-spot region: the COCCON Paris campaign 2015, Atmos. Chem. Phys., 19, 3271-3285, https://doi.org/10.5194/acp-19-3271-2019, 2019.

Wunch, D., Wennberg, P. O., Toon, G. C., Keppel-Aleks, G., and Yavin, Y. G.: Emissions of greenhouse gases from a North American megacity, Geophys. Res. Lett., 36, L15810, https://doi.org/10.1029/2009GL039825, 2009.

Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W., Sherlock, V., and Wennberg, P. O.: The total carbon column observing network, Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences, 369, 2087–2112, https://doi.org/10.1098/rsta.2010.0240, 2011.