Authors Response to Reviewer's Reports

“Black carbon aerosol reductions during COVID-19 confinement quantified by aircraft measurements over Europe” by Ovid Oktavian Krüger et al.

We thank the reviewers for their insightful and constructive feedback. We implemented the feedback into a revised version of the manuscript. Please find our answers to the reviewers' questions below, accompanied by a marked-up manuscript version.

Reviewer Comment from Referee #1, 17 Jan 2022

Citation: https://doi.org/10.5194/acp-2021-1100-RC1

This manuscript is a straightforward and relatively brief analysis of vertical profiles of airborne black carbon (BC) measurements made over western Europe in July 2017 prior to the COVID-19 pandemic and in May and June 2020 during the "lockdown", or "confinements", of much personal and industrial activity. The intent is to show that lower BC mass concentrations (Mbc) in 2020 relative to 2017 can be attributed to differences in emissions due to the lockdown, which varied between countries in western Europe. To account for varying meteorology, the ECHAM/MESSy model is used to simulate the Mbc for each of the sampling periods. The HALO aircraft was "flown" through model space to calculate median profiles that can be directly compared to those measured by the aircraft. Median vertical profiles measured in 2020 were about 47% lower than in 2017 (when integrated vertically to get a columnar loading value). According to the model, only about 7% of this difference was attributable to meteorological/transport differences, while 40% of the difference was attributable to reduced emissions during the shutdown period in 2020 (and a slight, long-term decreasing trend in BC emissions).

The manuscript is well written and, as I said, rather straightforward. It will be of interest to the general public and policymakers, but is likely not to be much of a surprise to atmospheric scientists. Emissions went down, so the atmospheric loading went down roughly proportionally. But documenting this is worthwhile, and I find the paper appropriate for publication in ACP with relatively minor revisions.

Author response: We thank Referee #1 for the revision of the manuscript and constructive suggestions. We addressed the individual comments below.

Major comments:

The structure of the manuscript is unusual for ACP, and appears more like a Nature or Science format. There is a quite short main text body that discusses the findings and leaves many questions unanswered, followed by a very extensive Appendix that provides the experimental details, modeling parameters, etc. I don't have a particular problem with this format, but initially I was wondering where all the details had gone to. I suggest that the authors add a brief paragraph near the front stating the structure of the paper, and that details of the measurements, modeling, and results will be found in the Appendix, perhaps even outlining the Appendix. As detailed below, there are some spots where more information needs to be given in a single sentence, with the appropriate section of the Appendix pointed out.

The manuscript is structured according to the ACP letter format guidelines. We appreciate the comment of reviewer 1 and added a sentence in the end of the introduction pointing the reader to the additional details in the appendix.

I would very much like to see the equivalent of Fig. 3, but for the model results, in either the main sectio or in the Appendix. A lot can be learned by looking at how well the model simulates the spatial pattern of in situ data. I'd be especially curious to see if the very large Mbc values found between 2 and 4 km in the data during 2017, from biomass burning transport, is simulated by the model.

We thank for this comment. We updated Fig. 3 accordingly and included it into the appendix as Fig. A10. We further included an arithmetic mean boundary layer height and terrain height below the flight track into this figure. As apparent in the modified figure, the large values for M_{BC} between 2 and 4 km from biomass burning emissions are captured by elevated modeled M_{BC}, however the magnitude tends to be underestimated in this region. We extended discussion in section A5 accordingly. Further we included one additional paragraph in the appendix for the methodology of the boundary layer height retrieval and terrain height (section A6).

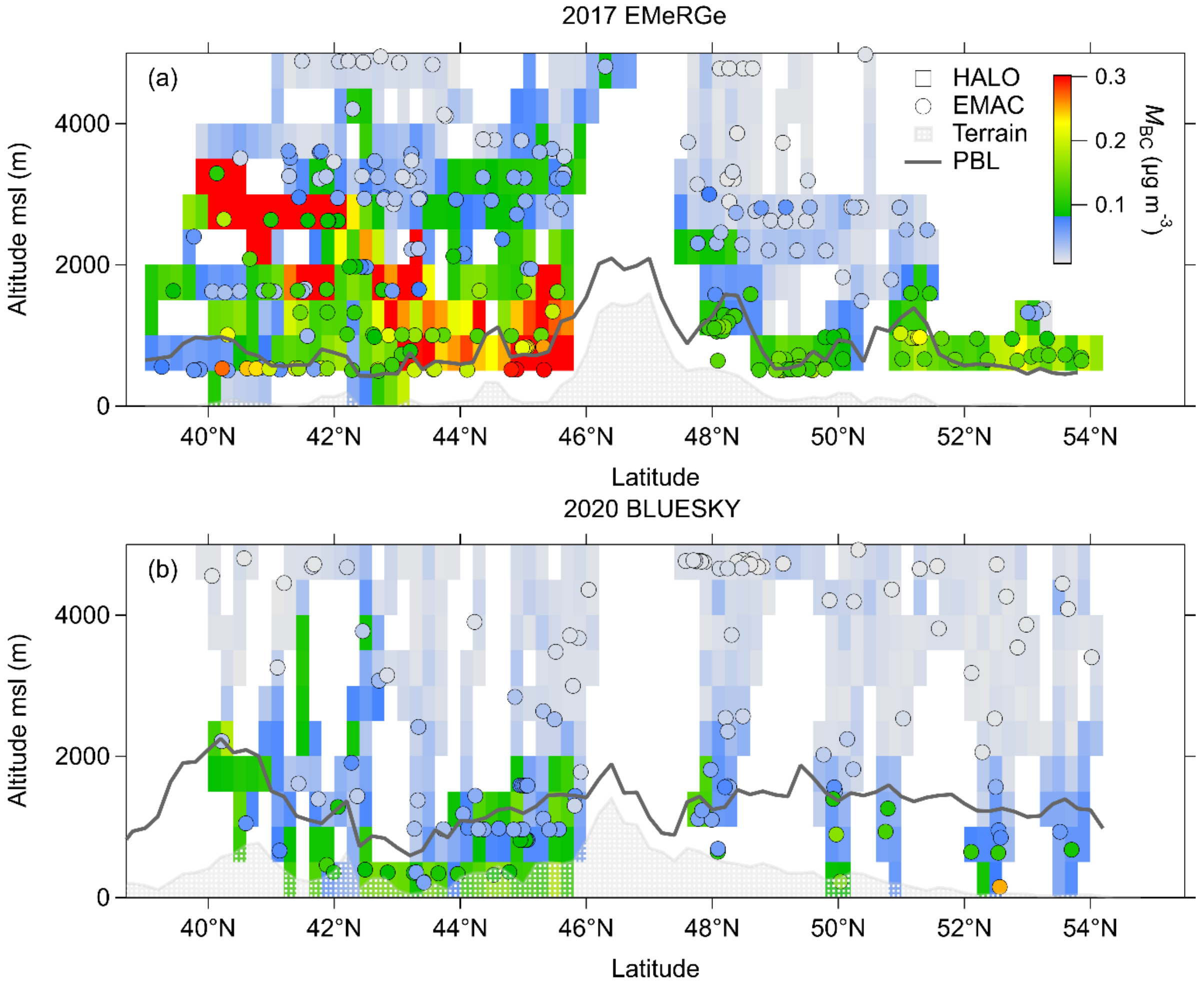

Fig. A10 as Fig. 3 with EMAC model results as round markers as well as arithmetic mean terrain height and arithmetic mean PBL height below flight track.

The authors need to discuss in the main text difference between free tropospheric measurements vs. the planetary boundary layer. This topic is brought up in Appendix A1, but I believe this needs to be emphasized more. The altitudes in Fig. 2 and 3 are not clearly defined; is the altitude above mean sea level, or is it above local terrain? It might make more sense to plot everything against altitude above ground level, which can be obtained using a digital elevation model database; there are several readily available. The average PBL height could also be shown, to help differentiate between locally/regionally emitted BC and that transported from long distance.

The arithmetic mean terrain and planetary boundary layer (PBL) height are now included in Fig. 3 as well as in Fig. A10. We decided to use the height above mean sea level for the summarizing vertical profiles and for the axis in Fig. 3. As shown in Fig. 3, most of the measurements were above locations with rather low elevation and using the height above sea level facilitates an easy comparison to previous studies and the EMeRGe EU and BLUESKY overview papers (e.g. Watson-Parris et al. 2019, Ding et al. 2019, Andrés Hernández et al. 2022, Voigt et al. 2022). Fig. 3 also shows, that we measured within and above the PBL. Also, the gradient in concentrations relative to the PBL is apparent in Fig. 3 for the measurements in Western Europe. Whereas M_{BC} in Southern Europe tends to be also lifted into higher altitudes. Some of these higher concentrations are due to contribution of transported biomass burning smoke. Potential other reasons for the larger M_{BC} concentrations above the PBL are upward transport triggered by topography as well as deep convection as transport pathway.

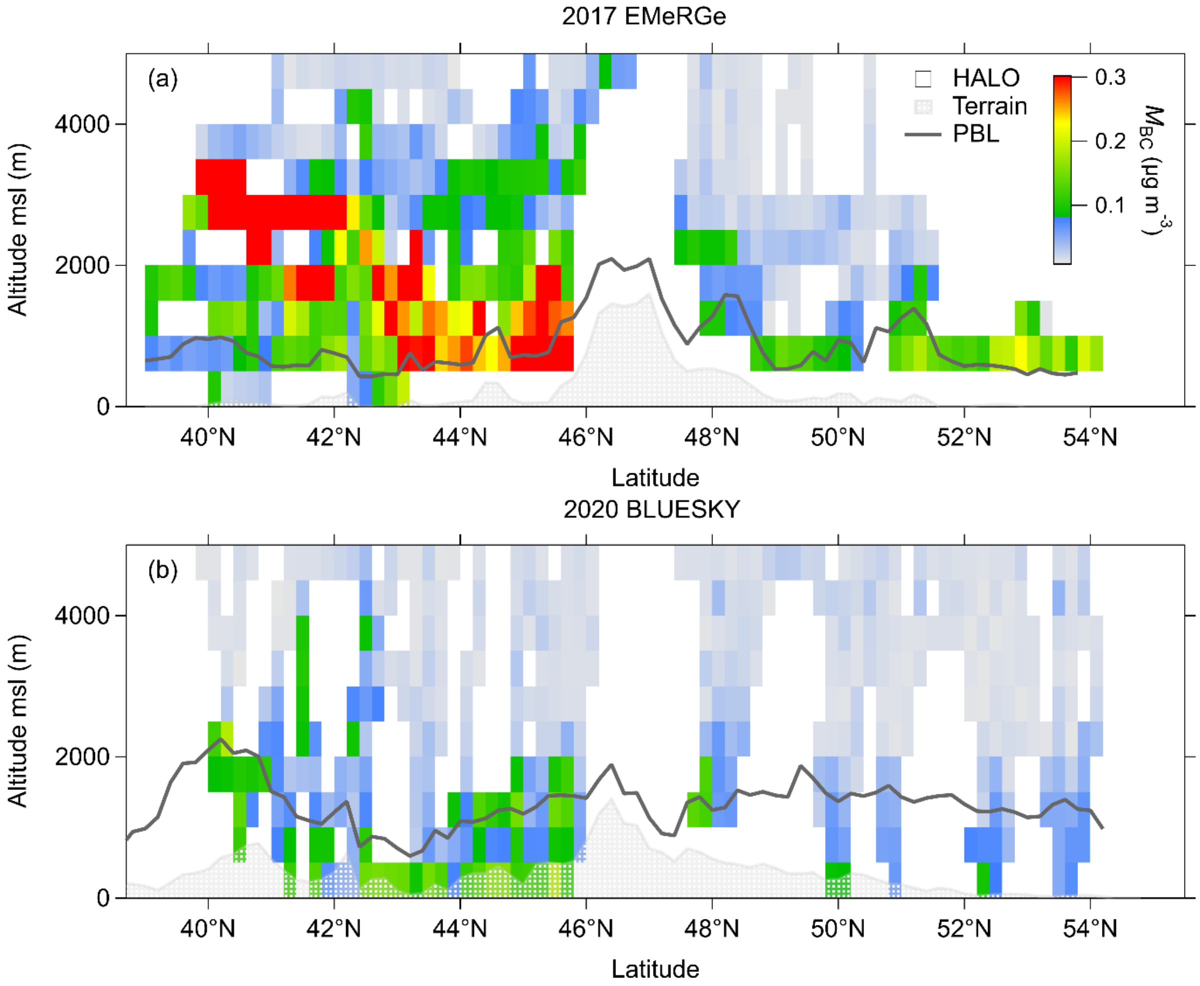

Fig. 3. Measured arithmetic mean of M_{BC} per 0.2 ° latitude times 500 m altitude bins with arithmetic mean terrain height and arithmetic mean PBL height below flight track.

Line 111: Can you define the "lockdown period"? Is May 2020 not in the lockdown? I see that this is well documented in the Appendix, but I think it would be helpful to show one of these graphs in the main text and clearly define what is mean by the lockdown period since it is used frequently in the main text.

There were several ways of response to minimize the spread of COVID-19 in early 2020. The responses with the largest impact on air pollution include closings of schools and workplaces, the cancellation of public events, stay-at-home requirements and relinquished restrictions on both international and internal travel (Hale et al. 2021). We extended the description of the lockdown period in section A2 as well linked in the introduction to the corresponding figure and section in the appendix and added a brief description of the strength of the lockdown confinements in line 36.

Minor Comments:

Figure 2: Is the y-axis altitude above sea level, or above the local surface?

We updated the figure captions and axis. Now it is altitude above mean sea level throughout the study.

Figure 2: You say solid lines represent "average" values. "Average" is not mathematically defined. Is this the geometric mean, the arithmetic mean, or some other type of average? I assume the arithmetic mean. In that case, it's interesting that the mean and median values are so different for the 2017 Mbc data, suggesting some strong transport events between 1500 and 3500 m that are driving the mean to much larger values than the median. (This ends up being discussed in the Appendix; a reference here to that section of the Appendix would be helpful.)

We updated the figure caption and the methods (section A8) stating that we use the arithmetic mean and referred to the further discussion of the wildfire smoke transport events during EMeRGe EU.

Figure 2: The caption says that Mbc is shown in panels A, B, C, and F, but it's shown in A, B, C, and D. Panel F shows the BC core diameter. The caption also says that panel E shows the measured "Mbc"; it should be "Nbc".

Thanks a lot, we correct that.

Lines 110-115: How much BC is emitted from heavy goods vehicles vs light duty vehicles? Did the lockdown affect them both the same? (Again, this appears later in the Appendix but should be discussed briefly here.)

A quantification of the emissions from heavy vs light duty vehicles is exceeding the scope of that study. To our knowledge, there is still a lack of real-world data on traffic emission factors for different fuel, engine and vehicle types (e.g. Peitzmeier et al. 2017 found for measurements in a medium sized German city EURO5 and EURO 6 limits for particle number concentration exceeded by a factor of 150 in real-world measurements compared to legal limits). Also, a shortcoming of regulation and thus of measurements of BC is an issue in Europe. BC is only indirect regulated by defined limits for particulate matter and particle number concentrations. Even more complex are real world emission factors for aged traffic pollution. Therefore, these measurements are missing even though their high relevance for air quality, health and climate issues.

The effect of the lockdown on traffic was not the same for light and heavy goods vehicles. Whereas the volume of traffic for light vehicles decreased by roughly 50 % during the lockdown in Germany, heavy goods vehicle traffic was only reduced by around 25 % (IW, 2020).

We extended the discussion of the fuel demand data from line 144 to include the above mentioned.

Line 119: what are "motor spirits"? Gasoline (petrol)? Or Diesel? Is kerosene different than Diesel? Or is this aviation (jet) fuel? Please clearly define your fuel terms, since there are substantial differences in usage of these terms across different countries.

Motor spirit is the term used by EUROSTAT for gasoline products. As pointed out kerosene is aviation fuel of kerosene type (not including gasoline type aviation fuels which only account for a very minor fraction of aviation fuels). Diesel differs from kerosene in its cetane number. We agree that this are not common terms and therefore, we extended the description of these quantities in section A11 and changed in the main text to a more common nomenclature with reference to the description in A11.

Line 124: Please define what is meant by "solid fuels". Coal, biomass, anything else?

We extended section A11 and added a brief description in the discussion (line 159). Solid fuel is defined by EUROSTAT as Hard coal including: Anthracite, Coking coal and other Bituminous coal; Brown coal including: Sub-bituminous coal and Lignite and Coal products including: Patent fuel, Coke oven coke, Gas coke, Coal tar and Brown coal briquettes (https://ec.europa.eu/eurostat/statistics-explained/index.php?oldid=449721}, last access 19.04.2022).

Line 138: By "general emission reductions" do you mean "reductions associated with long-term trends"?

Yes. We updated the description to specify more accurately “general emission reductions”.

Line 144: The sentence beginning "In particular. . ." is not a complete sentence.

We have adjusted the sentence accordingly.

Figure A8: What are the units on the x-axis? This graph has a gray background that is different from all the others.

We updated the figure accordingly. The unit on the x-axis is W m^{-2}.

Figure A9: If panels A and B were placed on the same scale we could more clearly see the reduction in Mbc during BlueSky as opposed to EMeRGe.

We updated the figure accordingly to make the comparison of both campaigns straighter forward.

Reviewer Comment from Referee #2, 07 Feb 2022

Citation: https://doi.org/10.5194/acp-2021-1100-RC2

This study evaluates the effect of lockdown on the BC emission using aircraft-measured vertical profiles and modelling adjustment. It is a concise and well-presented study and contains substantial valuable work. Although the conclusion itself is not particularly exciting, the dataset is valuable and would worth publishing after addressing the following points.

Author response: We thank Referee #2 for the revision of the manuscript and constructive suggestions. We addressed the individual comments below.

My main concern is how the 40% reduction of the overall emissions has been derived. How robust this value is. Why only a single value to adjust on the old inventory to apply for all over the regions in Europe. What is the criterion, has the comparison been performed with the measurement in the boundary layer or free troposphere to derive this conclusion? Would some sensitivity tests about this 40% be required?

We thank the reviewer for that valuable comment. We extended our analysis accordingly to provide an adequate answer to this question.

The BC concentrations we obtained in this study represent background concentrations for European continental air masses. Since European countries are rather small, and atmospheric background concertation are strongly interacting across national borders, a comparison of single states seems not as the adequate measure for our data set. Also, the high speed of the aircraft relative to the ground (around 800 km h^{-1}) suggest a coarse spatial resolution analysis as more meaningful. Therefore, we did a separation at 47 °N to evaluate the regional sensitivity. The threshold for the separation is based on a rough definition of the Alps (based on our measurement flight tracks e.g. Fig. 1 and Fig. 3). Also, the higher concentrations in Southern Europe compared to the region North of 47 °N are supporting the selected threshold. We included all measurement data to achieve a better statistical basis for our analysis. This is different to the approach described in section A8 for the vertical profiles of the measurements where we only included data 30 seconds before and after an EMAC model output. To facilitate a consistent analysis, we also extended the measurement data set for the general comparison shown in Fig. 2. Due to the slight changes in data used for this analysis we find the general reduction to be 41 % for adjusted vertically integrated M_{BC} burden. We updated the values in the Results section accordingly.

The separation for North and South of the Alps is included in Fig. A6. We find a good agreement with the overall reductions shown in Fig. 2. The reductions in M_{BC} between 2017 and 2020 in Southern Europe is 37 %, consistent to the corresponding value for Northern Europe of 38 %. We included an additional paragraph in Sec. 2 presenting these findings.

The reduction we find in this study is based on our aircraft in-situ measurements and the EMAC model serves to facilitate a direct comparison accounting for the multivariate set of impact factors on atmospheric aerosol measurements (e.g. meteorology, seasonality in emissions, wildfire emissions) and different flight pattern. We show in Figs. A11 and A12 the good agreement between model and measured values for synoptic data as well as for M_{BC}. The additional model run with 40 % reduced M_{BC} serves as a general check for the measurement-based findings. Thus, further model sensitivity studies do not seem to be adequate for our in-situ measurement focused study. A model improvement and tests against measurement results, however, is indeed a very interesting topic and we encourage case studies making use of our measurement data doing so.

Our measurements represent data from both inside the planetary boundary layer (PBL) and aloft in the lower free troposphere (see Fig. 3). As apparent in Fig. 3 M_{BC} is not strictly confined to the PBL, however, a strong altitude dependent gradient exists. This vertical distribution of BC agrees well with observations reported by Ding et al. 2019 for the European atmosphere. Therefore, we limited our comparison to the lower 5 km (msl) of the atmosphere and compared vertically integrated M_{BC} burdens, where the center of mass is within and close to the PBL.

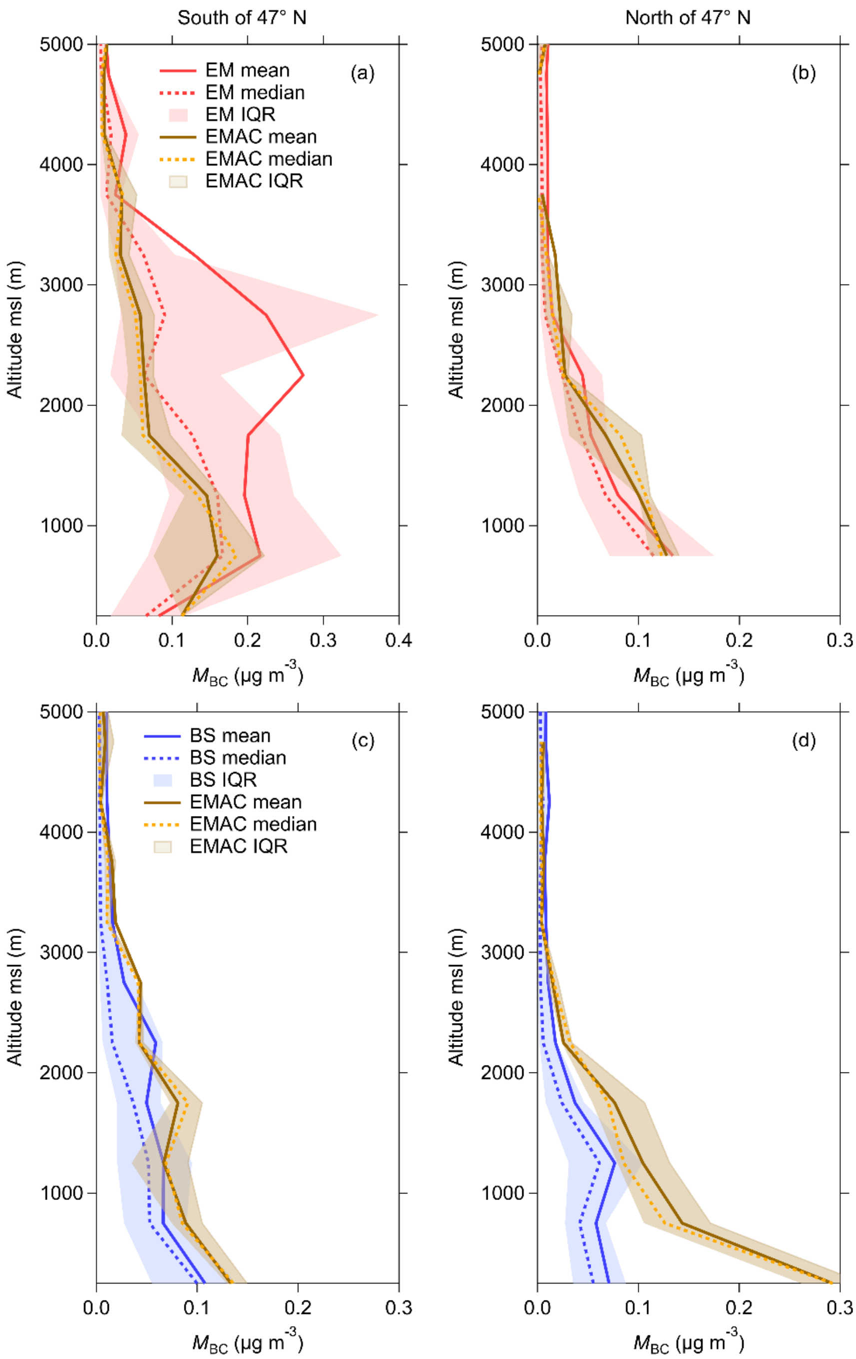

Fig. A6: Vertical Profiles North and South of 47°N. Note that x-axis in (a) ranges from 0 to 0.4 µg m^{-3}.

Other points:

1) One important point I think is from the plot, it seems the surface BC concentration has not been reduced significantly, what is the reason for this?

This can be explained by the differences in flight tracks. While there was the great opportunity to perform measurement flights near major emission centers as well as in background regions in very low altitudes during the BLUESKY campaign (due to almost no air traffic and great cooperation of the European air traffic control) these flight tracks were not possible during the EMeRGe EU campaign due to dense traffic. We extended the description of the flight tracks and campaign in section A1. These differences, however, are considered in the EMAC simulations (e.g. see Fig. A6 d) and thus included in the adjusted vertical profiles.

The large discrepancy between model expectation and actual measurements in Fig. A6 d can be explained by the several low approaches in the flight pattern (see section A1) which allowed us to probe near surface PBL pollution at Northern European major airports (e.g. Frankfurt Main, Amsterdam Schiphol and Berlin Tegel). Especially in these areas of highly frequented airports close to major pollutions centers the reduced anthropogenic emissions are apparent. Other than these very specific and targeted maneuvers we had only limited near surface flights in the Northern region. In Southern Europe we were able to also perform measurement flights near surface above the Mediterranean and Atlantic Ocean beside some low approaches in e.g. in Madrid, Barcelona, Bordeaux, Rome and Marseille. Thus, our data in the Southern region includes more background measurements.

It is important to note, that the lowest altitude bin for measurements North of 47 °N were not included in the reduction calculations, since there were no comparable measurements during the EMeRGe EU campaign in 2017 performed (see Fig. A6 b and Fig. 3). As stated in section A9 we only included data in the comparison where we have values during both HALO aircraft campaigns.

In the vertical distribution of M_{BC} in Southern Europe during summer 2017 (Fig. A6 a) the mean exceeds the upper quartile. This shows nicely the biomass burning smoke influencing a relatively small fraction of the measurements but causing a strong shift of the mean concentration, whereas the median is robust against event like concentration peaks.

2) It would be useful to indicate the mean boundary layer height during flights, as you were mainly focusing on the pollution reduction in the boundary layer.

We included the average boundary layer height as well as the terrain height in Fig. 3.

3) Line 75-80, has biomass burning significantly changed between both years? You mentioned the high-altitude was more influenced by biomass burning, but the later discussions have not mentioned it. It may be useful to simply show the fire points to imply how they have changed.

We included two figures showing the MODIS Fire Radiative Power (FRP) for the campaign periods, illustrating the differences in fire activity. These figures are presented in the results section.

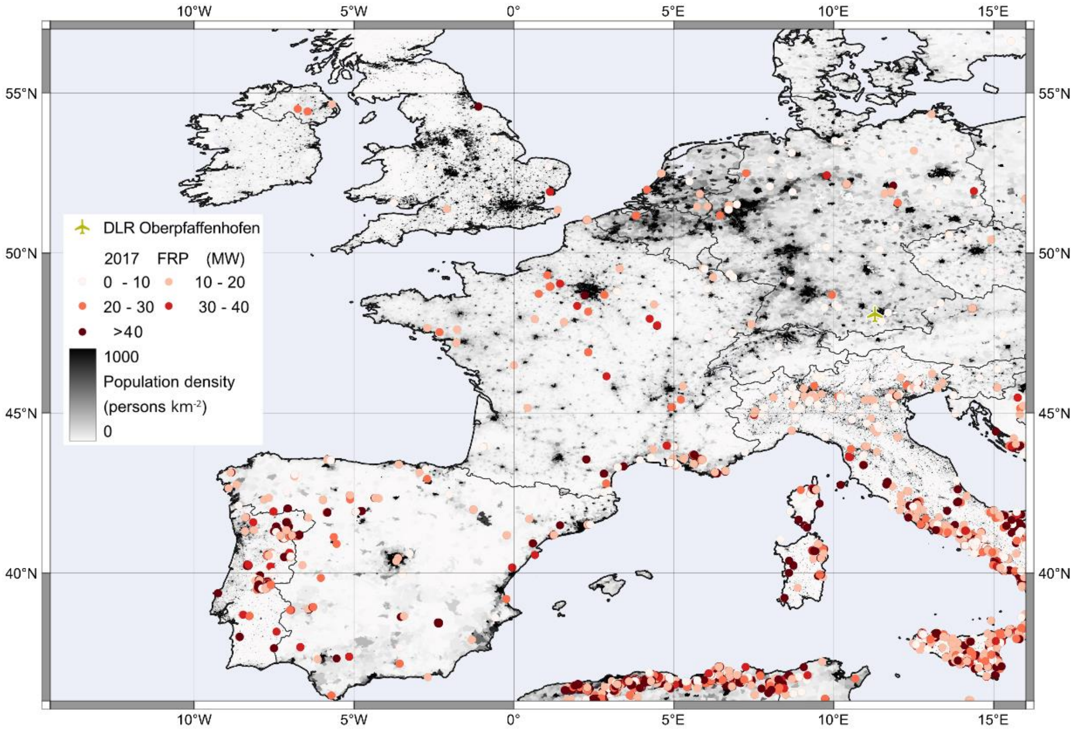

Fig. A13: Fire radiative power in megawatts (MW) for measurement period in 2017.

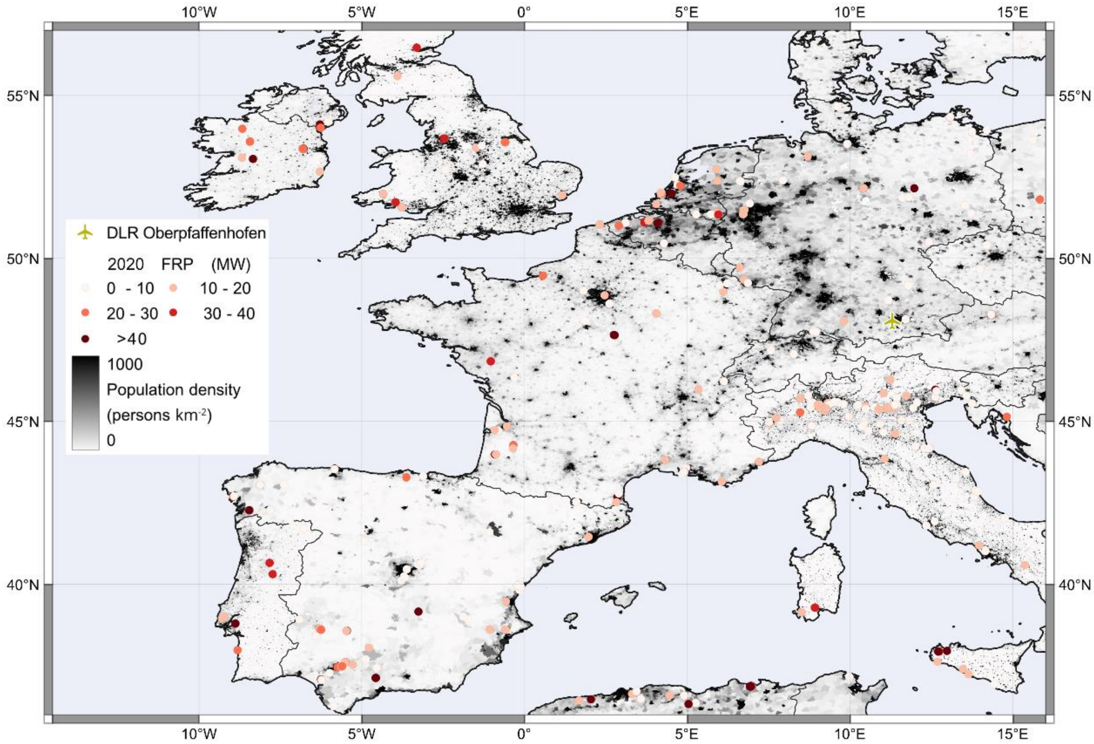

Fig. A14: Fire radiative power in megawatts (MW) for measurement period in 2020.

4) How about the other fractions besides the meteorological and emissions, as the sum is not really close to 100%.

We are sorry for causing confusion in the description of receiving the actual reductions due to the COVID-19 lockdown. We reformulated the paragraph in the results stating the reduction. However, the sum of adjusted 2020 measurements and reductions is 100 % of the 2017 measurements. We extended the description of the calculation for the emission reduction in section A7 as well as in the results section.

5) Have you considered the diurnal variation of BC mass loading the boundary layer, i.e. the flight time in the day.

No, this is not included into the data analysis, however all measurement flights took place exclusively during daytime. Further we extended the discussion about our measurements since the median concentrations which are the basis for our conclusions represent atmospheric background concentrations in Europe (Querol et al. 2013) where no significant diurnal cycling is expected. We extended the description of the measurements and that there is no significant diurnal variance expected for the median vertical profile in section A1.

6) In Figure 2, I would suggest adding the inventory information in the plot legend, not just mentioning it in the caption. Has "grey and solid line" been shown in both Fig. 2a and d? The concentration normalized by non-emission factors between 2017 and 2020 needs to be more clearly clarified. Have you modelled the 2020 case using exactly the same met data with 2017? Have the flight path been set the same. I would suggesting a rather simple and clear plot to show the procedures step by step how the met influence has bene neutralized.

We updated the figure caption. Further indicated already in the figure caption where to find details on our approach to facilitate the data comparison.

We also extended and clarified the description of our approach to normalize for factors different than emission reductions in the results section and section A7. We only calculated an adjusted vertical profile for BLUESKY (i.e. 2020, figure 2 d). The approach for normalizing the vertical profile is based on the difference between the expected BC concentrations from the two initial model runs.

The meteorological data used in the model runs nudging is the ERA-5 re-analysis data. The approach to make the measurements comparable is by using the same emission inventories (Hoesly et al. 2017). This allows us to account for the impact of factors other than reductions in anthropogenic BC emissions.

The flight path in the different EMAC model runs are not the same We used the model as a tool to account for exactly these differences (e.g. flight path, transport pattern, removal processes and other BC sources than anthropogenic emissions). By comparing the model results for 2017 and 2020 from the initial model run we receive what BC concentration we could have expected, given no emission changes took place. This concentration we used in the following to account for all factors different than emission changes to facilitate a meaningful comparison of the aircraft in-situ measurement data. We also extended the descriptions in section A1 and the results (from line 71) to clarify this approach.

7) The fact is that most of the light tracks have not been overlapped, some discussions are required to explain the reasoning to allow for this comparison.

We extended the description of the flight tracks in section A1 as well as the results (from line 71) and A7 where we describe our approach to facilitate a direct comparison of the two aircraft data sets (see also reply to question 6 above). The comparison of aircraft data and in general of atmospheric measurements is an issue we address with the use of the EMAC model. Since the transport of pollutants from emission sources is strongly depended on the synoptic situation, even an overlapping of flight tracks would not allow a meaningful comparison of the measurements. A better understanding of the transport pathways, losses and emission burdens by means of EMAC chemistry-transport model simulations facilities the comparability of the different measurement flights.

8) It would be useful to discuss the point that reduced BC concentration corresponded with the reduction of BC core size (maybe due to reduced chance of coagulation between BC particles) and its potential implications. This statement can be made by comparing the BC core size with some regions which are significantly influenced by anthropogenic emissions and how this has been related to BC mass concentrations (Liu et al., 2020; Ding et al., 2019). It would be helpful to comment whether precipitations had affected the BC core size.

This is a very interesting reasoning, we included into our discussion. However, we do not expect coagulation to have a significant influence on the BC size distribution over Europe since the median N_{BC} is over the whole vertical profile reported for both campaigns is rather low (below 100 cm^{-3}) which tends to be too low for significant coagulation to play a role. Therefore, the influence of biomass burning is likely a factor influencing the rBC core size distributions in 2017 stronger than in 2020. The larger rBC core size diameters are reported by e.g. Schwarz et al., 2008 and Liu et al., 2014.

Differences in coagulation growth tend to have an impact on particle size, however the differences we found are rather small and the second impact of coagulation is on particle number concentration. Whereas coagulation increases the mean BC core diameter by less than a factor two (assuming spherical particles) the number concentration is linear affected by a factor of two. Due to the good agreement between reductions in mass concentration and number concentration, combined with rather marginal differences in BC core diameters, we do not expect coagulation to show a significant difference for lockdown to normal conditions. This can be due to general lower BC concentrations in Europe compared to China or global wildfire hot spot regions and thus a lower probability of particle coagulation in the polluted lower troposphere.

Ohata et al., 2016 found precipitation predominantly removing accumulation mode BC particles. The vertical distribution of BC core diameters provided in figure 2, however, shows that both the median BC core diameter as well as the mean is well centered in the accumulation mode. Liu et al. 2020 showed a decrease in the residual BC core diameters subsequent to scavenging within the PBL. During BLUESKY we observed smaller rBC core diameter, however, during May/June and July there are no significant differences in precipitation for Europe in contrast to the observations reported by Liu et al., for dry winter time and wet summer time in the Beijing region. We also expect a shift to smaller rBC core sizes subsequent to precipitation. Anyhow, our data can unfortunately not serve to adequate quantify the importance of wet scavenging on the black carbon background concentration or size distributions in Europe. To answer that question, experiments have to designed differently (e.g. measurements of the same air masses before and after precipitation event or cloud droplet and rain droplet residual sampling and analysis). Also, a profound quantification or measurement of precipitation did not take place during our measurements.

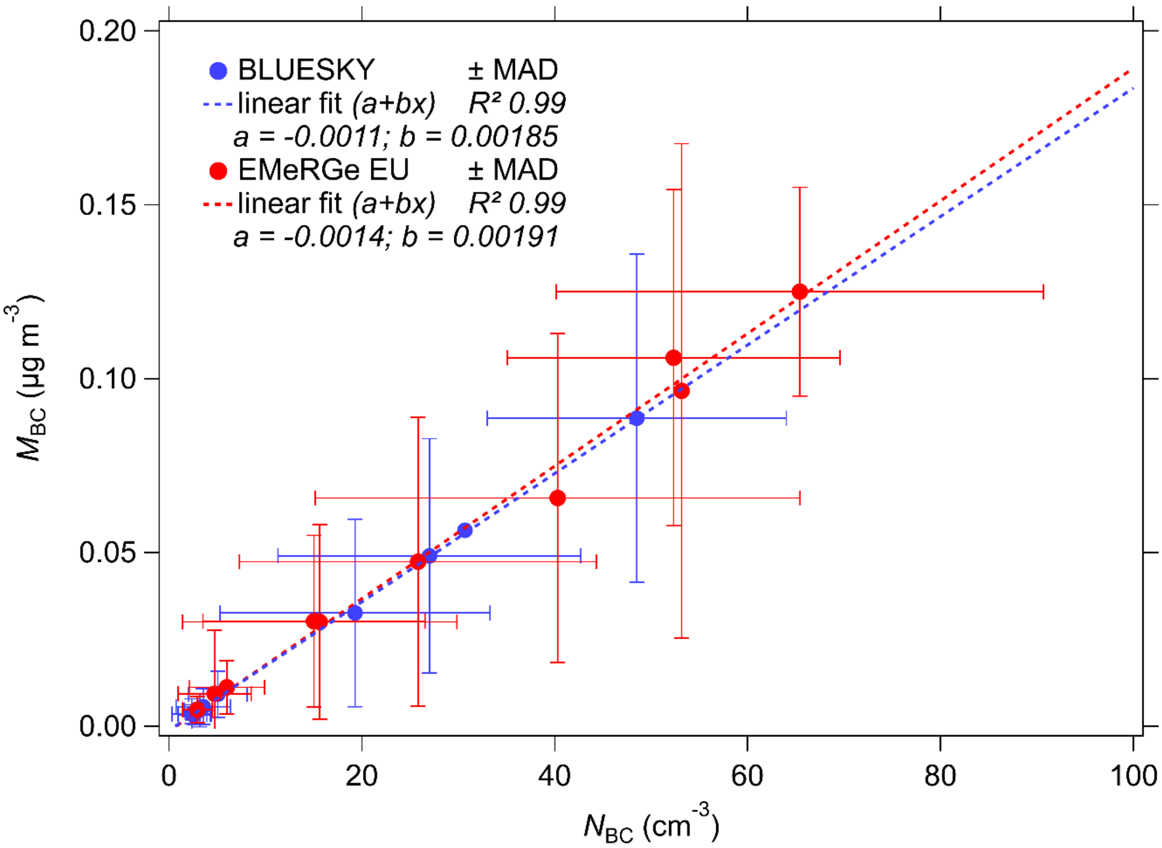

Fig. A15: M_{BC} vs N_{BC} for the vertical distribution. The linear ratio between M_{BC} and N_{BC} is makes us confident that no large differences in coagulation during both campaigns were taking place.

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