We thank the reviewer for her/his comments. Here they are reported (in bold) with our replies. Following the comments of referee #1 and these comments, we have decided to change the title of the manuscript, to fit better with the content of the manuscript: 'Numerical simulation of the impact of COVID-19 lockdown on tropospheric composition and aerosol radiative forcing in Europe'"

General comments

• Introduction: The literature review tends to omit primary references and focuses on recent work, with quite a few imprecise descriptions of key processes (that this long list of expert authors could easily address).

Following the comments of referee #1 the introduction has been revised.

• Methods: The description of the model and of the setup of the simulations is insufficient, which affects reproducibility and the ability to interpret the results.

We have followed the suggestion of the referee (see specific comments) to improve this section. The model description is now more precise and detailed.

• Context : manuscript entirely ignores an international model intercomparison project on this very subject, CovidMIP, which has been published months before submission of this manuscript (Jones et al, GRL, 2021). (Disclaimer: I am not involved in CovidMIP.) Clearly, the results of this study should be put into the available context but beyond that, it needs to be clear what additional insights are gained other than the focus on a specific area. The availability of a dedicated aircraft campaign provides ample opportunity to do this but is currently not exploited beyond a baseline evaluation of the model.

We thank the referee for pointing out this very important intercomparison project, which we indeed have overlooked. We have added this reference in the introduction and we have summarized the outcome of this project. Nevertheless, we argue that our study only partially overlaps with CovidMIP, which does not include a comparison with observations. It is thus difficult to judge, whether the series of model results presented in CovidMIP are reflecting the "real" state of the atmosphere during the lockdown. Here, we us a state-of-the-art CCM to reproduce the observed state of the atmosphere (to the extend that is possible) to base a qualified estimate of radiative impact on this (evaluated) model. We also note that a number of manuscripts is in preparation or have been submitted to a special issue in ACP(D) "BLUESKY atmospheric composition measurements by aircraft during the COVID-19 lockdown in spring 2020" (e.g. Nussbaumer et al., 2021; Hamryszczak et al., 2022). These manuscripts will include aspects of modelling and thus additional insights will be presented.

• Analysis : The interpretation of the results tends to be quite speculative and is held back by not tracing the perturbations through the full chain of relevant processes and by a lack of dedicated sensitivity studies necessary to back up some of the interpretation of the results.

We believe that we were not clear enough in our manuscript. More than 15 different sensitivity simulations were performed for our study, while only the basic simulations are presented, so to not overload the reader with information. We are hesitant to add a description and discussion on all these simulations simulations to the main text, as this would only detract from the main focus and make the manuscript less readable. Therefore we prefer to rephrase parts of the manuscript, also following the suggestions of the referee in the specific comments.

Specific comments

We sincerely thank the referee for reading the manuscript accurately and pointing out the text which was incorrect or not detailed enough. We believe that the manuscript has largely improved now that these specific comments have been taken into account.

• Where possible, please use primary references. For example, the trade-off between GHG and aerosols was not discovered in 2019 or the dependence of forcing on surface albedo is not something new from the Belloin et al. (2020) paper...

We consider the work of Bellouin et al. (2020) an excellent review on the topic (with detailed references therein) with up-to-date estimates, and we would like to keep the citation. Never-theless, we further added a citation of Shindell et al. (2013). For the GHG-aerosols trade-off we cite the IPCC report, which offers a comprehensive review of the state-of-the-art for this topic (Myhre et al., 2013).

• Line 73: "The cloud albedo effect can be enhanced by the Twomey effect" is very confusing as they tend to be used synonymously. I do not understand what is meant here.

The introduction has been fully rewritten and this sentence removed (see also replies to referee #1).

• Line 79: "The same effect arises in aircraft flight tracks..." claims analogy between ship-tracks (albedo enhancement of existing clouds via Twomey effect or LWP increase) and the formation contrails but this is really not the same.

The introduction has been fully rewritten and this sentence removed (see also replies to referee #1).

• Line 88: Cloud lifetime effect is introduced without giving credit to Albrecht and treated as a fact, rather than a long-standing (and often disputed) hypothesis. The cited references are fairly outdated.

The introduction has been fully rewritten and this sentence removed (see also replies to referee #1).

• Line 132: "The aerosol-cloud interactions are based on the aerosol microphysics parameterization of Pringle et al. (2010) including aerosol aging and the continuous calculation of aerosol number concentration depending on the mass mixing ratio and mixing state." This is not a description of aerosol-cloud interactions but of the underlying aerosol microphysics. Which key processes are represented and how? To name a few: updraft velocities, activation, the link from activated particles to CDNC (in particular in presence of existing droplets), the effect of CDNC on cloud microphysical (through autoconversion/accretion) and radiative properties.

We agree that the sentence was not precise and the scheme of Pringle et al. (2010) indeed describes the aerosol microphysics. We modified the sentence by adding more information on the aerosol representation (we added information on cloud formation and the aerosol-cloud

interactions in the next reply (L135)). The new text is the following: "Atmospheric aerosols are described via a two-moment aerosol scheme, which predicts number concentration and mass mixing ratio of the aerosol modes (Pringle et al., 2010). This scheme takes into account various physical-chemical processes of aerosols, such as coagulation, aging, condensation, and also the gas-aerosol partitioning (Fountoukis and Nenes, 2007)."

- Line 135: "Large-scale cloud formations and prognostic variables depending on cloud microphysical processes follow the work of Lohmann et al. (2007); Lohmann and Hoose (2009); Bacer et al. (2018)." This seems unlikely as Lohmann et al describe a cloud microphysics scheme and you seem to refer to the cloud fraction scheme (which presumably is Sundquist but this is not described at all).
 - The sentence was not precise, because Lohmann et al. indeed describe a two-moment cloud scheme and we did not specify the cloud cover scheme used in the simulations (the one of Sundqvist et al., 1989). We changed this sentence and we wrote more details on the cloud representation and the interplay with aerosols as following: "Convective cloud processes are accounted for using the framework of Tost et al. (2006), based on the convection schemes of Tiedtke (1989) and Nordeng (1994). Convective cloud microphysics does not take into account the influence of aerosols on liquid droplet or ice formation processes and is solely based on temperature and vertical velocity. In EMAC, the vertical velocity is given by the sum of the grid mean vertical velocity and the turbulent contribution (Brinkop and Roeckner, 1995), thus one single updraught velocity is used for the whole grid cell. Large-scale stratiform clouds are described by the CLOUD submodel, which, in the setup applied here, uses a two-moment cloud microphysics scheme for cloud droplets and ice crystals (Lohmann et al., 1999; Lohmann and Kärcher, 2002; Lohmann et al., 2007). and solves the prognostic equations for specific humidity, liquid cloud mixing ratio, ice cloud mixing ratio, cloud droplet number concentration (CDNC), and ice crystal number concentration (ICNC). Cloud droplet formation is computed by the "unified activation framework", an advanced physically based parameterization (Kumar et al., 2009; Karydis et al., 2011) that combines the κ -Köher theory (Petters and Kreidenweis, 2007) for the activation of soluble aerosols with the Frenkel-Halsey-Hill adsorption activation theory (Kumar et al., 2009) for the droplet activation due to water adsorption onto insoluble aerosols. Ice formation occurs via homogeneous ice nucleation following the parameterization of Barahona and Nenes (2009) and heterogeneous ice nucleation of insoluble dust, insoluble black carbon, and glassy organics following Phillips et al. (2013). In the cirrus regime $(T \leq 238.15K)$, the effect of pre-existing ice crystals and the competition for the available water vapor between homogeneous and heterogeneous ice nucleation mechanisms are taken into account (Bacer et al., 2018). Given the high contribution of instantaneous freezing (Bacer et al., 2021), the ICNC in the cirrus regime was modified according to Neubauer et al. (2019) in order to reduce the artificial homogeneous freezing of dry aerosol particles independent of availability of water vapor. Other microphysical processes related to cloud droplets and ice crystals, like phase transitions, auto conversion, aggregation, accretion, evaporation, melting, are also taken into account by the CLOUD submodel. The cloud cover is computed diagnostically with the scheme of Sundqvist et al. (1989), which is based on the grid-mean relative humidity."
- Line 140...: "We performed four simulations ... without cloud-aerosol interaction" casually refers to simulations performed without cloud-aerosol interactions. This is not a trivial exercise using a two-moment cloud microphysics as clouds

droplet number concentrations are prognostic and, if decoupled from aerosols, need to be initialised somehow (and the base-state will affect the results due to inherent nonlinearities) but no details are given on how this is done.

We apologize for the information missing in the manuscript. We augmented the manuscript with the following lines: "The model setup without cloud-aerosol interactions uses the original ECHAM5 cloud microphysical scheme (Lohmann and Roeckner, 1996) and a statistical cloud cover scheme including prognostic equations for the distribution moments (Sundqvist et al., 1989). Details on the cloud microphysical scheme can be found in Roeckner et al. (2003, and references therein)."

• It is difficult to put the results from this study into the wider context, such as AeroCom or CovidMIP, without a summary of the of ERFari and ERFaci from PD-PI simulations. As we currently have limited constraints on ERFaci from observations, it is important to know where the model lies in the ERF uncertainty range e.g. from IPCC AR6 or the Bellouin et al (2020) assessment. This should be included and discussed either in the methods or results section.

An analysis of direct and indirect effects with a model set-up very similar to the one used in our study can be found in Lelieveld et al. (2019). With regard to direct and indirect effect (and their spatial distribution) we refer to Fig. 5 of their supplement. We added the following lines in the revised manuscript: "Following the work of Lelieveld et al. (2019), the EMAC model in a setup very similar to ours, simulates a radiative forcing global mean of all anthropogenic aerosols at TOA (top of the atmosphere) of $-0.46 \pm 0.01Wm^{-2}$ and $-1.2 \pm 0.1Wm^{-2}$ for ERF_{ari} and ERF_{ari}+ERF_{aci}, respectively. At BOA (bottom of the atmosphere) the model simulates $-1.6 \pm 0.02W/m^{-2}$ and $-2.1 \pm 0.1W/m^{-2}$ for ERF_{ari} and ERF_{ari}+ERF_{aci}, respectively."

• Line 164: Measurement cut-offs are quoted but it is not clear if and how these are applied to the model size distributions in the evaluation. Are they explicitly applied for each component, how are internal mixtures dealt with – or are they ignored? And if they are, how would this affect the results?

The measurement cut-offs are applied to the model results when these are compared to the observations, in order to compare the same size range. More precisely they are applied for each individual component, to obtain a meaningful comparison for each instrument/component. The aerosols in the model are considered to be internally mixed (see reference to Pringle et al. (2010)). We clarified this point in the revised manuscript.

• Figure 2: The evaluation of O3, CO, NO is looking very good. Has there been any calibration/tuning during the setup of the simulations or is this out of the box?

The model was not tuned for these trace gases. The results are obtained from the simulation results as described in Sect. 2.1.

• Line 206: Here and later it is hypothesized that the underestimation of SO2 (and later on SO4) is due to representation of transport from the boundary layer or from the stratosphere to the upper troposphere or due to model short-comings within the stratospheric aerosol chemistry – but no further sensitivity studies are conducted to underpin this hypothesis.

We understand the concern of the referee and we agree that this should be investigated in more detail. As both, sulfate and SO_2 , are simultaneously underestimated, this points to some model deficit, and in the manuscript, we indicate that this is most probably (due to strong sulfate and ozone correlation) of stratospheric origin (e.g. due to volcanic eruption, line 243). In fact, at the altitudes where sulfate was measured, ozone has practically only stratospheric origin, and the strong correlation implies same sources for the sulfate. Nevertheless, despite various sensitivity simulations with the inclusion of the various eruptions prior to the field campaign, the underestimation is still present. Therefore, a more comprehensive study is needed to fully understand the model deficiencies, which is ongoing. Although the sulfur dioxide emissions could be increased to match the observational data, this would not help to solve the real problem. More detailed work, with additional observational datasets (e.g. satellite observations, field campaigns at similar and higher altitudes) is necessary to fully investigate this issue, and this is clearly outside the scope of our study. In addition, also instrumentation issues at higher altitudes cannot be ruled out: in fact, on a recent comparison with the same model and identical emissions total, the sulfate profiles compared to measurements from various aircraft campaigns were quite satisfactory (Pozzer et al., 2022).

• Line 217: "The measured black carbon (BC) concentrations are captured well by the model close to the surface, while the observational variability is underestimated at high altitudes." This seems to neglect the significant bias – it looks like median concentrations are almost an order of magnitude out in the upper troposphere? This section also needs to explicitly caution (not only in the caption) that you switched plots from a linear to a log scale

Indeed the referee is correct, with the observed median being much lower than the simulated one. However, it must be stressed that the measurement variability is extremely large, showing a very skewed distribution of the measurements at such altitudes. A detailed analysis of the black carbon concentration simulated with an almost identical model setup can be found in Krüger et al. (2022), and therefore we prefer not go into detail in our manuscript. We added to the these informations to the manuscript.

• Line 235: "A single factor causing the model underestimation of BC and sulfate aerosol concentrations in the upper troposphere, e.g. a localized plume of pollution, is judged unlikely, as BC and SO2 do not correlate" I am not sure I follow the logic here. This would be true if both would stem from the same source but for plumes arising from entirely different sources it seems plausible to find low correlations – while biases may still be affected by the same process such as a common transport or removal process.

The referee is completely right. In fact, the sentence should read: "A single emission source causing the model underestimation of BC and sulfate aerosol concentrations in the upper troposphere, is judged unlikely, as BC and SO_4^{2-} do not correlate".

• Line 269: "It must be stressed however, that those relative changes in 270 the upper troposphere, although significant, have a very minor impact on most trace gas budgets, due to their low mixing ratios at these altitudes." Mixing ratio is conserved under vertical displacement – you probably mean low concentrations (due to exponential pressure decrease)?

We have removed the sentence for clarity.

• Figure 3 5: are concentrations normalized to STP (needs to be clear in the caption)?

The concentration are not normalized to STP. We have add this information to the manuscript.

• I am missing an effort to use interesting measurement data and the evaluation to provide some constraint or context for the following analysis of aerosol radiative effects. As a minimum it would be helpful to analyse if the simulated change in response to the emission perturbations are larger than the underlying model biases (which would add trust) or not (which would add less trust).

We thank the referee for pointing this out. Although two sections are dedicated to the evaluation (3.1 and 3.2) and one (4.1) to the change in mixing ratios/concentrations, we did not spell out if "the simulated change in response to the emission perturbations are larger than the underlying model biases". From a detail analysis, the perturbation observed from the COVID-19 lockdown is, for the tracer, generally larger than the model bias to the observations. On the other side, for the aerosols components, the perturbation is smaller than the bias observed (see Figs.2,3 and 5). Nevertheless, we argue that the bias in the comparison would be present in all model simulations, therefore canceling out once the effects on radiation are estimated by subtracting the model results. To make the reader however aware of the issue, the following text has been added to the revised manuscript: "While the changes in CO and NO can be considered significant and representative of the real atmospheric changes (due to the low bias at all tropospheric levels between model results and observations), changes in the aerosol components should be considered with caution, as these are generally smaller than the bias between the model results and the observations."

• This section (and subsequent use) should stick to well defined nomenclature of aerosol forcing as used by IPCC, i.e., be clear what is RF, what is ERF, what is ari and what is aci. This also means that ERF should include SW and LW and it is not clear why the analysis is restricted to SW only.

Indeed the referee is here completely right, and we apologize for having ignored the IPCC nomenclature. First of all, the forcings included in our study are, following the Myhre et al. (2013) definitions, RF (radiative forcing) and not IRF (Instantaneous Radiative Forcing) or ERF (Effective Radiative Forcing). This is due to the fact that the tropospheric temperature is somewhat constrained by the prescribed SST/SIC (consistent with the nudging data) and the temperature nudging (patterns only), while the stratosphere is dynamically free. Moreover, for the applied model setup the "meteorology" of the BASE and RED simulations are binary identical. It must be stressed that the EMAC setup used here was in Quasi-Chemistry Transport Model (Q-CTM Deckert et al., 2011), and therefore we cannot derive IRF or ERF as in a fully coupled model setup. We have clarified this in the manuscript. Furthermore, the RF estimated are the effect of the COVD-19 lockdown against a baseline scenario (i.e. Business as usual), and therefore does not represent the radiative effect of all anthropogenic aerosols.

For the sake of completeness, we have also added the RF_{aci} and RF_{ari} to the manuscript (see Tab.1), although we decided to keep the analysis of the SW radiation as it is, as, to our knowledge, these are the most meaningful results, as the LW radiation is constrained due to the prescribed SST/SIC and the temperature nudging. Furthermore, following the definitions in Myhre et al. (2013) and Bellouin et al. (2020), RF_{ari} and RF_{aci} cover both

Table 1: RF_{ari} and RF_{aci} at the top of atmosphere (TOA) over Europe for May for the lockdown against a baseline scenario.

	$\mathrm{RF}_{\mathrm{ari}}$	$\mathrm{RF}_{\mathrm{aci}}$
TOA	0.08 ± 0.03	0.19 ± 0.92

the solar (shortwave, SW) and terrestrial (longwave, LW) parts of the electromagnetic spectrum. Therefore we decided to not use this nomenclature when writing only of the shortwave radiation, so to avoid possible misunderstanding.

We would like also to point out that even in the CovidMIP analysis performed by Jones et al. (2021), only the shortwave radiation is investigated (aerosol radiation interaction), without any reference to total radiation or aerosol cloud interactions, as this was the only statistically significant anomaly identified. Analogously to us, Jones et al. (2021) also avoided the nomenclature RF_{aci} and RF_{ari} , as only the shortwave radiation were taken into account (see above).

• This section would be much more intuitive if it followed the actual chain of processes from aerosol properties, through aerosol radiative properties (AOD, AAOD) all the way to the radiative fluxes.

We thank the referee for the suggestion. Nevertheless, we prefer to keep the structure as it is and extend as possible the missing part, so to facilitate a comparison with the original manuscript.

• Line 289: Fluxes defined at what level?

This is mentioned in line 294: ground level.

• Line 298: "our radiative effect of all aerosols in our RED simulation for May is of 3.33 ± 1.36 Wm2, which is close to their value of 2.3 Wm2." The definition of "radiative effect" is entirely unclear here.

The referee is right. The sentence is not clear and based on erroneous data. We have removed the sentence as it was not essential for the manuscript.

• Line 303: "the total absorption (clear sky) was decreased by 0.064 ± 0.053 Wm2, with slightly more than one third of this caused by the BC decrease" How do you know?

As mentioned before, various sensitivity studies were performed to proof our assertions. In this case we performed a simulation equal to STD (now BASE), but only with decreased BC emissions according to the lockdown. Similar simulations were performed performed for SO_2 and NO, to analyze the radiative effects of reducing individual components. Although the individual perturbation results are not summing up linearly, the radiation impact of the individual reduction during lockdown provided a fully closed budget within their uncertainty ranges. We added to the revised manuscript the text in line 303: "was decreased by $0.06 \pm$ $0.05Wm^{-2}$. Based on an additional sensitivity simulations, in which only individual emissions (i.e. of BC, SO_2 , NO) have been reduced, we found that slightly more than one third of this reduction is caused by the BC decrease".



Figure 1: Simulated differences in heating rates with a reduction of all emissions (black) and a reduction of BC emissions only (red).

• Line 309: "The decreased heating (for the entire column but mostly 310 at the surface) is due to the reduced absorption by BC". Again, how do you know? We thank the referee for pointing this out. As mentioned in the previous answer, this is based on the results of additional sensitivity simulations to evaluate the importance of BC. We checked again the results and, as shown in Fig.1, we found that the decrease of BC is responsible for roughly 40% of the decrease of the shortwave heating at the surface. We have corrected this in the revised manuscript.

• Line 314 / Fig 8. What is N – how is it defined? Is it total CN without size-cut off?

In contrast to the comparison with the observations (which had a cut off), N is here the total aerosol number concentration simulated by the model. Analogously, also ICNC and CDNC are without size cut off. We added this information to the revised manuscript.

• Line 323: "these differences (in N, CDNC, ICNC) can be directly connected to the reduced air traffic present during the lockdown (REDCLOUD)" UTLS aerosol tends to be dominated by nucleation (not sure if this included in N or not as it is not defined) so the attribution to aircraft is ambiguous. I am really missing a process-based analysis here from emission to CN to CCN/INP to CDNC/ICNC – and from the model description it is not even clear what processes could actually affect CDNC/ICNC. Likewise, the attribution to specific emission sectors should not be based on speculation – it would be trivial to run a simulation with and without the aircraft only emission reductions to make this point.

As part of the sensitivity simulations performed, we have performed a simulation, in which the BASECLOUD (formerly STDCLOUD simulation) was repeated, but with reduced aircraft emissions. In Fig. 2 the results are shown, compared to the simulation BASECLOUD. It is shown that the reduction of aircraft emissions caused a strong decrease in the particle



Figure 2: Vertical profiles of the monthly mean aerosol number concentration from simulation BASECLOUD (blue), REDCLOUD(RED) and a sensitivity simulation based on the same but with only reduced aircraft emissions (green) for May 2020 over Europe. The grey line depicts the relative difference between REDLCOUD and the sensitivity simulation. The grey area denotes the spatial and temporal standard deviation of the relative differences.

numbers in the upper troposphere. We calculated, from our model results, that more than 90% of the reduction of particle number concentrations at 200-300 hPa are attributable to the missing aircraft emissions at these altitude, while less than 10% are caused by reduction of other emissions, proving that aircraft emissions reduction is mostly responsible for the strong decrease in particle number at 200-300 hPa.

• Line 334: There is really very limited point to quote RF/ERF with three significant figures in the presence of significant noise and uncertainty.

We agree with the referee and we modified the figures to two significant digits.

• Line 338: "This confirms the importance of the cloud-aerosol interaction, as mentioned by Hong et al. (2016), Gasparini and Lohmann (2016) and Myhre et al. (2013)." This is of course not new but the split is highly model dependent so this should be discussed early on (as suggested above).

We have removed this line and included this discussion in the introduction.

• Line 345: "Nevertheless, problems remain regarding stratosphere-troposphere transport, especially of volcanic influence, which resulted in systematically underestimated SO and SO2of stratospheric origin, and a consequent overestimation of NO3 (which substitutes the underestimated sulfate in ammonium salts) in the upper troposphere." This could be true but no results are provided to underpin this, nor is a reference given that shows this.

This has been proven in many studies and is of common knowledge (e.g., Seinfeld and Pandis, 2008; Bauer et al., 2007; Xu and Penner, 2012). These reference were added to the revised manuscript.

• Line 354: "With reduced emissions, the model simulates a lower number concentration of aerosols; this reduction is located at an altitude too high to effectively influence the cold cirrus clouds" From the model description it is entirely unclear if or how "aerosols" could actually affect cirrus in this setup.

We included information about the influence of aerosols on cloud formation in the model description (section 2.1, see replies to L132 and L135) and we changed the sentence at L354 to: "With reduced emissions, the model simulates a lower number concentration of aerosols between 300 and 50 hPa; this reduction is located at an altitude too high to influence the cloud droplet formation (Karydis et al., 2017) and heterogeneous ice nucleation from black carbon and dust; glassy organics freeze at these altitudes, but their contribution is totally negligible in comparison with homogeneous nucleation (Bacer et al., 2021)."

• Line 356: "The analysis of the indirect aerosol effect did not give any conclusive results, due to the large variability in the calculations caused by the short duration of the lockdown "experiment". I agree but you also make the argument that these "effects" dominate the overall result, so this suggests that this could be noise?

We partially disagree with the referee: these effect dominates only for the SW radiation at the TOA. At the surface, the aerosol cloud interaction effect on SW radiation is comparable to the direct effect, although with much larger variability. On the other hand, the referee is right in mentioning that the changes of cloud properties add noise to the system, which masks the overall effect. Possibly, an ensemble of simulations (or a much longer simulation) could reduce the variability, although this is outside the scope of our study.

• At a minimum, the data going into the plots should be deposited in an open access archive.

The observational data and the model results are available on the HALO (High Altitude Long RAnge research aircraft) database (https://halo-db.pa.op.dlr.de), upon sign of data protocol. This was included in the revised manuscript.

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