

Dear editor, here a item-by-item response of referee #1 comments. As we believe only referee #1 specific points were missing, we have added only those to this document. We listed each single change with also the line numbers of the new manuscript.

For safety we attached also the differences between the updated manuscript and the on-line version at the end of this document.

Best regards,

Andrea Pozzer on behalf of all authors.

The title of this manuscript suggests the authors intend to explain aerosol radiative effects during the early stages of the pandemic with reduced emissions. However, it is not clear to me that they have made a convincing case to justify the title after reading the manuscript. In particular, in the manuscript, they describe a model (nudged by meteorology) with four simulation runs targeting the period of interest. They say their model does a "reasonable" job in general, but this assertion needs further contextualization and reasoning. Overall, a reader like me is left wondering what the authors are actually trying to tell in this manuscript: Is it the "unique" measurement campaign, a "unique" model they developed to match these measurements, or fundamental new progress and understanding about aerosol radiative effects?

A chemical-climate model of such complexity as the one used in our study cannot perfectly simulate observations, not even in specified dynamics mode. On the other hand, a chemistry transport model or air quality model, which might be better suited to simulate the observations, cannot easily be used for radiative impact calculations. We use a CCM (instead of a CTM) to base an estimate of radiative impact (of reduced emissions during the World-wide COVID lock-down) on observations of atmospheric trace gases and aerosols. The agreement between our model simulations and the observations from a tailored measurement campaign using research aircraft is comparably good for most of the trace gases and aerosols, especially when compared to other intercomparison based on other model setup (e.g. Jöckel et al., 2010; Pozzer et al., 2022). We believe that our evaluation is quite comprehensive, although room for further analysis is obviously present. In order to avoid giving the false impression, we agree to change the title of the article, to fit better the content of the manuscript. The new title "Numerical simulation of the impact of COVID-19 lockdown on tropospheric composition and aerosol radiative forcing in Europe" reflects the content of the manuscript, for which we simulated the "unique" measurement campaign during the COVID-19 lockdown.

Major points

- **What is the purpose of this paper? Is it the model only or the associated knowledge/insight produced by this model in concert with the measurements? There is a clear disconnect between the title and introduction on the one hand and the rest of the manuscript on the other. As an example, there is a lengthy discussion of aerosol effects in the introduction, yet it is not clear how anything in the rest of the paper fills any of the many gaps in our understanding of aerosol-cloud interactions. The authors should consider shortening the introduction (pointing the reader to further material) and instead focus on what they actually address.**

We thank the referee for this suggestion. We have reduced the introduction. In addition, as the referee pointed out, we decide to change the title of the manuscript, to better reflect the content of the manuscript: "Numerical simulation of the impact of COVID-19 lockdown on tropospheric composition and aerosol radiative forcing in Europe". To our knowledge, no

studies on European lockdown have been performed for the entire troposphere, by evaluating model predictions with the observation in the entire tropospheric column. Following the comments of the referee #2, we have instead extended the model description to provide more detailed information on the methodology.

- **The model evaluation is incomplete at best, perhaps quite weak. It can also be circular at times. The authors use a nudging technique whereby they anchor the model to some meteorology, then they evaluate the model by comparing some model outputs to aforementioned meteorology. Is that an accurate reading? Shouldn't they be the same by definition? More context and thorough explanation is needed here.**

This is a classical misunderstanding, at least partly because we did not provide sufficient information about our model setup: we apply a CCM in “specified dynamics” (“nudged”) mode, which is established by Newtonian relaxation of the prognostic variables divergence, vorticity, temperature (without global mean), and logarithm of surface pressure. This “nudging” is applied in spectral space by low normal mode insertion, i.e. on the synoptic scale only, in order to nudge the meteorology of the CCM towards the “observed” (indeed analysed or re-analysed) meteorology. Due to the scale limit of this procedure, a CCM in “specified dynamics” mode is still fundamentally different from a CTM, since the nudged CCM develops its own physics on sub-synoptic scales. Therefore, it is required and reasonable to compare the simulated meteorology with the available in-situ observations despite the nudging, as described in line 173. For an in-depth discussion on the Newtonian relaxation technique applied in spectral space, we refer to Jeuken et al. (1996) and Löffler et al. (2016). In our study we show that the agreement between the nudged physical state of the model atmosphere and the observations is good, despite some deviation on specific humidity (a quantity that is NOT nudged). We will clarify this point in the revised version.

- **In general, aerosol indirect effects are challenging and any work purporting to make progress in this field should be scrutinized. So please be precise and forthcoming about what this work actually brings to this field. Again, it is important and timely; so this is not to dismiss this work, but please be as precise as you could to contextualize your work.**

Indeed, our study does not focus on conceptual advancements or the development of new methodologies. We rather apply a state-of-the-art model to reproduce (by numerical simulation) data observed during the BLUESKY campaign, to further base a qualified estimate of radiative impact (namely of the reduced emissions). We believe our model results to be particularly robust, as the comparison with the observations shows a very good agreement at lower levels, where the highest concentration of aerosols is present. As mentioned above, for clarification, we decided to change the title of the manuscript to focus more on the impact of the world-wide COVID-19 lockdown in Europe.

Minor comments

- **While you are free to make up your own definitions, it is often not a good idea to make up acronyms that have other meanings in popular culture or other fields. For example, “STD” refers to sexually transmitted diseases in general, and in this manuscript, it refers to “standard” or “business as usual” — the authors should**

consider unifying their approach here: They use "STD," "business as usual," and "baseline" throughout the manuscript. One would suffice, preferably the last one, "baseline."

Thanks for pointing this out. We changed STD to BASE.

- **In many places there are added sentences that add no value to the text. For example, line 172 could be deleted; the first part of line 189 could also be deleted; lines 105–108 are unnecessary; the majority of line 157 can go as well. More on these in the list of technical comments below.**

We have followed some of the suggestions of the referee (e.g., line 105–108). In other cases (e.g. line 172), we decided to keep the sentence as this is needed to reference the figures and the table.

List of technical/specific comments:

- **COVID-19 takes a dash, not en or em dash.**

We have used only dash in the revised manuscript

- **Line 2: reads awkwardly "through direct... and indirectly," better use rephrase to use "directly ... and indirectly"**

Line has been modified following the suggestion as " Aerosols influence the Earth's energy balance directly by modifying the radiation transfer and indirectly by altering the cloud microphysics."

- **Line 4: delete "Here"**

The "Here" has been deleted

- **Line 9: delete "a somewhat"**

The "a somewhat" has been deleted

- **Line 10: delete "which could have ... campaign"**

The "which could have influenced the campaign" has been deleted

- **Line 10–11: replace "a business as usual scenario" with "the baseline"**

We have replace all the "business as usual scenario" with "baseline" in all the text, i.e. line 6 , line 10, line 35, line 57, line 86, line 152, line 256, line 283, line 299, line 313, line 331, label Figure 7, line 322, label Table 2.

- **Line 16–17: reads unclearly, maybe write: "ice crystal concentration, cloud droplet number concentration, and effect..."**

We have change the sentence following the suggestion and now reads " Impacts on ice crystal concentrations, cloud droplet number concentrations,and effective crystal radii are found to be negligible. "

- **Line 19: "millions of years of life expectancy" — not sure what the cited items say, but last time I checked, life expectancy refers to one person's life expectancy and so summing the whole planet's life expectancies to make a point is both**

unscientific and clumsy. I will leave it up to you to decide, but hopefully you will decide to keep the convention.

We have rephrase the sentence (line 2s1921 with) “ They have large impact on human health [...]”

- **Line 24: replace ”here” with ”hereafter”**

We have replaced “here” with “hereafter”

- **Line 27: by citing many works on air pollution and not citing a single work on climate effects, you’re positioning yourself as the only paper addressing this. First, that’s wrong because you’re not making a convincing case here anyway about climate effects per se. Second, there are many studies about the climate effects of the lockdown. Could you please cite them and contextualize how your work differs from them?**

We have removed the sentence “and climatic effects, on which we will focus in this work.”, as now we have new reference on climate studies, also based on the reply to referee #. In line 36-40 we added the sentence “ Many works are present in the literature that investigate the climatic effect of COVID-19 lockdown (e.g. Lee et al., 2021; Forster et al., 2020; Gettelman et al., 2021). Of particular importance is the CovidMIP intercomparison project, where 12 global chemistry climate model were used to investigate the impact of COVID-19 lockdown on the radiation (Jones et al., 2021; Lamboll et al., 2021), with special focus on aerosol-radiation interaction. ”

- **Line 35: replace ”business as usual” with ”baseline”**

We have replace all the “business as usual scenario” with “baseline” in all the text, i.e. line 6 , line 10, line 35, line 57, line 86, line 152, line 256, line 283, line 299, line 313, line 331, label Figure 7, line 322, label Table 2.

- **Line 37: remove ”, as will be ...”**

The sentence has been removed.

- **Line 43: ”wavelength of the radiation” — last time I checked this was more or less constant or basically unchanging during the lockdown, so what gives? Why do you have it here? It seems you’re implying that it is changing...**

The sentence is in our point of view correct: absorption and scattering of aerosols depends on the wavelenght of the radiation (Seinfeld and Pandis, 2008).

- **Line 50: last word, ”May” — which May? May 2020?**

The “2020” has been added in line 54.

- **Line 55: ”trigger several indirect effects” — awkward phrasing**

The “trigger several indirect effects” has been removed and replaced with “have several indirect effects”.

- **Line 56: ”alter cloud properties” should be better phrased, maybe ”can potentially alter cloud properties” or something similar**

We have replaced “alter cloud properties” with “can potentially alter cloud properties”

- **Line 64: avoid using two symbols after each other, use the word "approximately" maybe.**

Line 64 has been removed, as part of the introduction has been rewritten based also on referee #2 comments.

- **Line 99: "unique" may be a stretch.**

The word "unique" has been removed (line 87), and now reads " We use the BLUESKY observational data set of trace gases and aerosols obtained during an aircraft measurement campaign in Europe during the COVID-19 lockdown in summer 2020 to evaluate the model results. "

- **Lines 105–108 should be deleted**

It has been removed following referee's suggestion.

- **Lines 140–149: please use something other than "STD" here.**

We changed the name to "BASE".

- **Line 150: Is the "binary identical dynamics" relevant to your case? If so, please say more about it here briefly.**

We have extended the description with the text "i.e. they reproduce numerically exactly the same dynamics", line 162.

- **Lines 157–158: "led by ... with the aim of" can be deleted, the interested reader can just go read Voigt et al 2021 if they want.**

The sentence has been removed.

- **Lines 164–165: not sure if 40 micron is correct, is it? Also doesn't "aerosol particle number concentrations" cover the previous parts (e.g. ORG)? So what's going on with this list?**

The numbers are correct: line 164 is 40 nm, while line 165 are 40 μm . The ORG refers only to the organic material present in the aerosols (as mass).

- **Line 169: "sampled online" was the model running on the aircraft? If not, this sentence is wrong**

We have rewritten the sentence substituting "online" with "runtime" to make it clearer: " For the comparison, the model output was sampled during runtime by the submodel S4D (Jöckel et al., 2016), following the flight tracks of the field campaign and with a time frequency of 5 minutes. "

- **Line 170: What was the time step of the model? Sampling at 5 minutes seems too frequent for these types of models. Did you simply interpolate from the model time step or what is going on here?**

The model set-up used in our work has a time-step of exactly 5 minutes. We added this information on the model description, in line 103 " [...] the model has a time step of 300 seconds [...] ".

- **”Results: Model evaluation”** — either use an overarching one **”Results”** section or just drop the word **”results”** from sections 3 and 4.

We have removed the “Results” word, keeping only “Model evaluation”.

- **Line 172 can go.**

The sentence has been removed.

- **Lines 173–178 can also go; you should generally address the model validation better and this paragraph doesn’t do you any service.**

Based on our reply to the “Major Points” above, we hope we clarified the importance of the temperature evaluation. We kept therefore the sentence as it is.

- **Lines 179–181: yes, exactly. So maybe a different evaluation is needed**

Based on our reply to the “Major Points” above, we hope we clarified why the dynamics was evaluated. We have rephrase the sentence (line 188) as “ Overall, the agreement between the meteorological variables from model and the one observed in the BLUESKY campaign indicates successful initialization and nudging of meteorological variables and that the meteorological conditions during the relevant time period are simulated adequately. ”

- **Line 182: ”is not surprising” to whom? You could be more precise here**

The sentence has been reformulated (line 192), clarifying why the deviations should be expected: “ As the model is not nudged in the stratosphere or boundary layer (the nudging coefficient is maximal in the free troposphere (Jöckel et al., 2006)), the deviation in the upper troposphere between model results and observational data are to be expected, due to the intrinsic model dynamics which deviates from the nudging data. ”

- **Line 189: ”Observed ... by the model” should go**

We have rewritten the sentence (lines 199-202) “ More than 94 % of simulated ozone (O_3) mixing ratios are within a factor of 2 of the observations (”PF2” value) and the normalized root mean squared error of 0.04 is low, with improvements from previous evaluation of the same model (Jöckel et al., 2016).”

- **Line 193: ”within a factor of two” — is that any good? If so, please explain. If not, also please explain. In general give more context to these ranges, otherwise they can be interpreted differently by different people. For example, a factor of two is really bad in my opinion...**

See answer to previous point: now we have clarified that the comparison to ozone observations is better than previous evaluation (e.g. Jöckel et al., 2016) of the same model.

- **Line 194: delete ”somewhat”**

The word “somewhat” has been removed

- **Add readable legends to Figure 2.**

We have modified the legend and modified the caption of the figure. Now it reads ”RED simulation” and ”Observations”.

- **Line 206: "hypothesize" — please say more. Can we test this hypothesis? If so, how?**

The sentence has been removed and text extended in lines 251-255, also following comments from referee #2.

- **Line 212: delete "The vertical ... reproduced (see Fig. 3)." Also "quantitatively" in what sense? Can you qualify that more if you want to keep it?**

The sentence has been extended explaining what we meant with "qualitatively" : "The vertical profile of the measured aerosol number concentration is qualitatively reproduced (see Fig. ??, with logarithmic scale in the x-axis), with a minimum at $\simeq 300$ hPa and a maximum at the surface."

- **Lines 223–226: Please either list these volcanoes of interest (obviously super important; you do list some of them later, e.g. Line 243) or don't leave vague language like this around. This could be an opening for you to improve the manuscript anyway**

The volcanoes of interested are listed in line 257. The sentence in the same line has also been reformulated : "We tested this by injecting high levels of SO_2 in the stratosphere in additional simulations, mimicking volcanic eruptions that had enough energy to reach the stratosphere, i.e. Raikoke (June) and Ulawun (June and August) in 2019 (see de Leeuw et al., 2021; Kloss et al., 2021)."

- **Line 232: "We conclude that" — can you give your reasoning to this conclusion? Is it a conclusion anyway or an observation at this stage?**

We have changed the words "We conclude that [...]" with "We can observe that [...]"

- **Figure 3 like Figure 2 (add legends)**

Legends have been changed, as Figure 2.

- **Figure 4: please make it bigger and clarify it.**

The figure is now bigger. The figure is explain in lines 246-251.

- **Line 250: "Results" again, see my above comment about "results"**

The "Results" word has been removed

- **Line 257: these are not "purely attributable" to differences in your model or are not well captured by it, correct?**

In this sentence we mentioned that the two model simulations BASE and RED differs ONLY for the emissions, and not for the dynamics. This implies that they experience, for example, the same advection. As results, the differences between these two simulation are ONLY caused by the different emissions, while all other process are identical.

We rephrase it to make it clearer (line 265): " As no difference in dynamics between RED and BASE simulations are present, any chemical differences between these simulations are purely attributable to the different emissions during the lockdown period, as these are the only changes between these two simulations, and the consequent different chemical regimes."

- **Line 266: "most tracer" should be "most tracers"**

The word "tracer" has been changed to "tracers" (line 280).

- **Lines 270–271: please elaborate more on this.**

- **Figure 5: add legends and make bigger**

The legend is present in the upper left panel.

- **Line 284: "monthly mean sulfate (and inorganic aerosols, not shown) and black" should be rephrased**

The sentence has been changed. " Compared to the baseline emissions, the monthly mean sulfate (and inorganic aerosols, not shown) and black carbon concentrations are reduced in all troposphere, with a strong relative reduction at the commercial flight level [...].".

- **Line 286: "lockdown scenario" please unify your naming.**

We changed the words "lockdown scenario" to " scenario with reduced emissions due to lockdown", analogously to line 153 (where the model simulations are described).

- **Line 299: these are not really close values, are they? They are within the range of error. What's the range of error for the second value btw?**

This sentence has been removed, following the discussion with referee #2.

- **Line 302: is that significant?**

Yes, it is significant at 95% confidence level (via t-test).

- **Line 335: "We should note" instead of "We should notice"**

The text has been changed to "We should note".

- **Line 360: Could you reflect on this range a little more? Seems insignificant and uncertain to a casual reader.**

We have extended the text with the following lines: " The differences between these studies, can partly be attributed to the applied methodologies and general difficulties in discriminating anthropogenic effects from interannual variability. Hence, a study which considers contrail and aerosol effects simultaneously, and covers a longer time period, is recommended to better attribute the causes of the observed changes ."

- **Code availability: Why list all these details about doing MOU and all that, can you just give the git repository link and tag/commit?**

Unfortunately, due to the contained legacy models, we are not allowed to host parts of the code open source or publicly accessible. Our GitLab repository is not open and a URL would be therefore meaningless. This section is standard and used in the sister journals GMD(D) as mentioned in the license section of the MESSy system (<https://www.messy-interface.org/>).

- **Data availability: Please make your data available, and refrain from "contact the author" stuff. It doesn't seem open...**

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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Impact ~~Numerical simulation of reduced emissions~~ the impact of COVID-19 lockdown on ~~direct tropospheric composition~~ and indirect aerosol radiative forcing during COVID-19 lockdown in Europe

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Abstract.

Aerosols influence the Earth's energy balance ~~through direct radiative effects~~ directly by modifying the radiation transfer and indirectly by altering the cloud microphysics. Anthropogenic aerosol emissions dropped considerably when the global ~~COVID-19~~ COVID-19 pandemic resulted in severe restraints on mobility, production, and public life in spring 2020. ~~Here~~

5 ~~we~~ We assess the effects of these reduced emissions on direct and indirect aerosol radiative forcing over Europe, excluding contributions from contrails. We simulate the atmospheric composition with the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model in a baseline (business as usual) and a reduced emission scenario. The model results are compared to aircraft observations from the BLUESKY aircraft campaign performed in May/June 2020 over Europe. The model agrees well with most of the observations, except for sulfur dioxide, particulate sulfate and nitrate in the upper troposphere, likely due to a

10 ~~somewhat~~ biased representation of stratospheric aerosol chemistry and missing information about volcanic eruptions ~~which could have influenced the campaign~~. The comparison with a ~~business-as-usual~~ baseline scenario shows that the largest relative

differences for tracers and aerosols are found in the upper troposphere, around the aircraft cruise altitude, due to the reduced aircraft emissions, while the largest absolute changes are present at the surface. We also find an increase in all sky shortwave radiation of 0.327 ± 0.105 ~~0.21 ± 0.05~~ Wm^{-2} at the surface in Europe for May 2020, solely attributable to the direct aerosol effect, which is dominated by decreased aerosol scattering of sunlight, followed by reduced aerosol absorption, caused by lower concentrations of inorganic and black carbon aerosols in the troposphere. A further increase in shortwave radiation from aerosol indirect effects was found to be much smaller than its variability. Impacts on ice ~~crystal~~ and crystal concentrations, cloud droplet number concentrations, and effective crystal radii are found to be negligible.

1 Introduction

Aerosols play a pivotal role in both air pollution and climate change. They ~~cause millions of years of lost life expectancy per year globally~~ have large impact on human health (Lelieveld et al., 2015, 2020), impose a negative (net) effective radiative forcing (Bellouin et al., 2020), and are a large source of uncertainty in climate change assessments. A reduction of the cooling effect by a decreased aerosol burden necessitates stronger reductions of greenhouse gases (GHGs) for a targeted net radiative forcing (~~Larson and Portmann, 2019~~) (Larson and Portmann, 2019; Myhre et al., 2013).

Owing to the central importance of aerosol particles, the reduced emissions resulting from drastic restrictions on mobility, industry and public life during the ~~COVID-19~~ COVID-19 "lockdowns" in early 2020 (~~here~~ hereafter referred to as "lockdown") (Barré et al., 2020; Evangeliou et al., 2021; Guevara et al., 2021; Le Quéré et al., 2020) sparked a plethora of publications on the subsequent effects on local, regional, and global air pollution (see, for instance, He et al., 2020; Liu et al., 2020; Petetin et al., 2020; Tobías et al., 2020; Venter et al., 2020; Mertens et al., 2021) ~~and climatic effects, on which we will focus in this work~~.

We recognise that reduced emissions during lockdown do not necessarily translate into improved air quality, as primary pollutants take part in a complex set of chemical processes, which need to be included in a thorough analysis (Kroll et al., 2020). For instance, although ozone was reported to be reduced in the free troposphere in the northern hemisphere (Steinbrecht et al., 2021), the reduced emissions of the nitrogen oxides NO and NO₂ led to an increase in ozone concentrations in urban locations, as an important short-term sink (reaction with NO) was reduced (e.g. Gkatzelis et al., 2021; Sicard et al., 2020; Mertens et al., 2021). This illustrates how the complex (photo-)chemistry and the nonlinearity of the underlying chemical system have to be described and analyzed within the framework of a dynamic atmospheric chemistry model. A chemistry climate model with appropriate chemistry furthermore enables a direct comparison of ~~business as usual~~ baseline and reduced emissions within the same synoptic background conditions, complementary to a purely observation-based approach. ~~Such model investigations are most effective when accompanied and guided by observational data, as will be done in the present study~~ Many works are present in the literature that investigate the climatic effect of COVID-19 lockdown (e.g. Lee et al., 2021; Forster et al., 2020; Gettelman et al., 2021). Of particular importance is the CovidMIP intercomparison project, where 12 global chemistry climate model were used to investigate the impact of COVID-19 lockdown on the radiation (Jones et al., 2021; Lamboll et al., 2021), with special focus on aerosol-radiation interaction.

45 The interaction of aerosols with radiation and their climatic impact can be categorized into two types: (i) direct effects by impact on radiation fluxes, and (ii) indirect effects through changes in cloud physical and optical properties.

The direct effects include absorption and scattering of electromagnetic waves, whereby aerosol particles, most prominently black carbon (BC), absorb incoming solar radiation, which leads to warming of the ambient air and decreases solar irradiance in the layers below. In addition, aerosols scatter incident radiation back to space, leading to a net cooling of the climate system on average. These processes depend on the size, shape and chemical composition of the aerosols and on the wavelength of the radiation. In addition, the net effect depends on the surface albedo (Yoon et al., 2019; Bellouin et al., 2020).

(Shindell et al., 2013; Yoon et al., 2019; Bellouin et al., 2020). The reduced emissions in spring 2020 are thus expected to affect aerosol radiative forcing. A reduction in the backscattering of solar radiation is expected to result in warming, which is offset by the anticipated cooling effect through a reduction of black carbon emissions, and the net effect may vary vertically and horizontally. For instance, Gettelman et al. (2021) reported a simulated net warming at the surface and in the lower troposphere in most regions, caused by enhanced insolation at the surface, and cooling in upper layers of the troposphere, due to reduced absorption by black carbon. They also determined a difference in the clear sky net shortwave (SW) flux at the top of the atmosphere (TOA) of up to 0.1 Wm^{-2} globally in May 2020 between simulations with and without reduced emissions, i.e. less outgoing SW radiation due to the lockdown. Complementing the analyses regarding these more immediate effects, Forster et al. (2020) estimate a short-term warming driven by a weakened aerosol cooling through reduced sulfur dioxide (SO_2) emissions, followed by a cooling of $0.010 \pm 0.005 \text{ K}$ by 2030 in reference to a baseline scenario.

~~Tracks of conducted flights during the BLUESKY campaign (16th May to 9th June 2020). Colors denote the aircraft, Faleon (blue) and HALO (red).~~

In addition to the aerosol direct effects on the radiation budget, aerosol particles can trigger have several indirect effects. Aerosol particles serve as cloud condensation nuclei and thus can potentially alter cloud properties, such as cloud albedo, cloud droplet number concentration, formation processes, precipitation and cloud lifetime (see, for instance, Bellouin et al., 2020; Christensen et al., 2020; Lohmann and Feichter, 2005; Twomey, 1959). In turn, clouds also affect aerosols. Clouds convert precursor gases into aerosol particles through heterogeneous chemistry (Ervens et al., 2011; Lelieveld and Heintzenberg, 1992; McMurry and Wilson, 1983) and, at the same time, remove aerosols and soluble gases from the atmosphere by precipitation ("wet deposition").

~~Clouds are classified based on their liquid water or ice content, since these characteristics determine the efficiency of reflection of solar radiation back to space (cloud albedo effect) and the absorption rate of longwave radiation (greenhouse effect), thus their impact on the Earth's radiation budget. Clouds consisting only of ice crystals (e.g. cirrus clouds) occur mostly in high altitudes at temperatures below the homogeneous freezing threshold of $\sim -35^\circ\text{C}$ (Pruppacher and Klett, 1997; Krämer and Seifert, 2008). Ice crystals (ICs) are formed either due to homogeneous or heterogeneous freezing depending on aerosol characteristics and number concentration, temperature, supersaturation and vertical air motion (Pruppacher and Klett, 1997). IC number densities in cirrus clouds are typically lower (Voigt et al., 2017) compared to clouds at lower altitude, and cirrus clouds are often optically thin and transmit shortwave radiation and absorb longwave radiation, resulting in an overall positive net radiative effect at the TOA (Baer et al., 2018, 2021; Gasparini et al., 2017). In contrast, mixed-phase clouds, consisting of ICs and cloud droplets,~~

80 form only due to heterogeneous nucleation at lower altitudes and temperatures higher than -35°C . These optically thick clouds reflect a comparably large amount of the incoming SW radiation, leading to a negative net radiative effect at the TOA (Chen et al., 2000). The cloud albedo effect can be enhanced by the Twomey effect (Twomey, 1959), whereby high number concentrations of CCN lead to a higher number of droplets of smaller size compared to clouds formed with lower number of CCN. Accordingly, clouds, containing more droplets of smaller sizes, reflect solar radiation more strongly than clouds
85 containing fewer droplets of larger size. This effect is especially important over the ocean, where the number of natural CCNs, compared to anthropogenic CCNs, is much lower than over land (Lohmann and Feichter, 2005). Ship emissions, including aerosol particles, create clouds with a much higher reflectivity (or albedo) than clouds formed by the less numerous natural CCNs outside of the ship tracks (Platnick et al., 2000). The same effect arises in aircraft flight tracks, where the emitted particles may act as ice nucleating particles, leading to contrail formation (Schumann et al., 2017) and an increase in cirrus
90 cloud formation in the upper troposphere (Boucher, 1999). Still, the magnitude of the aviation soot effect on the radiation budget remains uncertain (Urbanek et al., 2018; Righi et al., 2021). Radiative forcing from aerosol cloud interactions is very challenging to quantify, and it is strongly model dependent (Hong et al., 2016; Gasparini and Lohmann, 2016; Myhre et al., 2013). Recently, satellite data have been used to quantify changes in clouds in regions with COVID-reduced airtraffic in 2020 (Quaas et al., 2021; Gettelman et al., 2021). With respect to contrails, Schumann et al. (2021a, b) find a substantial reduction of contrail
95 cirrus optical thickness and radiative forcing during the lockdown period.

The modification of cloud cover resulting from differing particle number concentrations has further effects on the lifetime of a cloud. An increase in smaller cloud particles makes the clouds more persistent and delays the occurrence of precipitation (referred to as cloud lifetime effect, Lohmann and Feichter, 2005). Increased cloud lifetime affects not only the timing but also the location of precipitation. The increased lifetime of clouds also extends the period over which a cloud can reflect radiation, thus adding to the Twomey effect. With increasing aerosol number concentration corresponding to a negative perturbation of the radiation budget, both effects contribute to cooling. Complex model as the one used by the CovidMIP, however, are most effective when accompanied by observational data, as the capability of the models to reproduce the real atmosphere is unclear, especially when the anthropogenic emissions are strongly perturbed as in the case of the COVID-19 lockdown. Furthermore, as the reduced emissions have different effect on the atmospheric composition depending also on the altitude,
100 the observational data should cover large regions of the troposphere. Despite the large presence of observations at the surface and at the TOA (Lohmann and Feichter, 2005). The number and size distribution of cloud nucleating particles can thus affect the extent of transmission of radiation during the COVID-19 lockdown (Gkatzelis et al., 2021), the free and upper troposphere presented comparably almost none in situ measurements against which the model could be validated. One notable exception is the BLUESKY field campaign (Voigt et al., 2021); from 16th May to 9th June 2020 in situ measurements of trace gases and
105 trace particles were conducted in the atmosphere over European urban areas and the North Atlantic flight corridor with the High Altitude and Long Range (HALO) research aircraft and a second research aircraft, Falcon (see Fig. 1 for flight paths). Comprehensive measurements of trace gases and aerosols composition were conducted, providing a unique set of observations that can be used to validate model results.

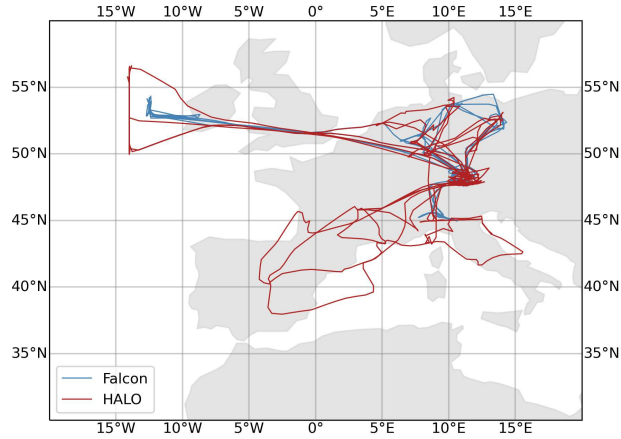


Figure 1. Tracks of conducted flights during the BLUESKY campaign (16th May to 9th June 2020). Colors denote the aircraft, Falcon (blue) and HALO (red).

Using an observation-guided model, the ~~COVID-19~~ COVID-19 lockdown provided an opportunity to examine how the climate system reacts to perturbations such as abruptly reduced air pollution emissions. The ~~COVID-19~~ COVID-19 lockdown may also serve to assess the impact of economic recovery with respect to climate change mitigation: for instance, Forster et al. (2020) show that investments aimed at a "green" opposed to a fossil-fueled recovery can reduce projected warming by 0.3 K by 2050, with only negligible contributions from the lockdown.

In the present study, we simulate the chemical composition of the atmosphere in Europe in spring 2020 under a reduced emission scenario and a ~~business-as-usual~~ baseline scenario with a state-of-the-art climate and chemistry simulation system, constraining atmospheric dynamics by reanalysis meteorological data. We use ~~a unique~~ the BLUESKY observational data set of trace gases and aerosols obtained during an aircraft measurement campaign in Europe during the ~~COVID-19~~ COVID-19 lockdown in summer 2020 to evaluate the model results. We then quantify the effects of the lockdown on radiative transfer in the atmosphere, particularly the change in shortwave fluxes and shortwave heating rates attributable to a reduced aerosol burden in Europe. Furthermore, we examine the impacts of the ~~lockdown-scenario~~ reduced emissions on cloud properties, including potential changes of the radiative forcing caused by indirect aerosol effects.

This paper is structured as follows. In Sect. 2 we describe the model together with an overview of the simulations performed (Sect. 2.1), as well as the observational data (Sect. 2.2). The model evaluation is presented in Sect. 3. We then investigate the impacts of the ~~reduced emissions during the 2020 COVID-19 lockdown on direct aerosol effects and indirect cloud-aerosol effects~~ (Sect. 4).

2 Data and methods

2.1 Model data

The ECHAM5/MESSy Atmospheric Chemistry (EMAC) model is a numerical chemistry and climate simulation system that includes submodels describing tropospheric and middle atmospheric processes and their interaction with oceans, land and human influences (Jöckel et al., 2016). It uses the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al., 2006).

For the present study we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.55.0) in T63L47MA-resolution, i.e. with a spherical truncation of T63 (corresponding to a quadratic Gaussian grid of approx. 1.8 by 1.8 degrees in latitude and longitude) with 47 vertical hybrid pressure levels up to 1 Pa. Roughly 22 levels are included in the troposphere, and the model has a time step of 300 seconds. The dynamics of the EMAC model has been weakly nudged (~~Jeuken et al., 1996; Jöckel et al., 2006~~) towards the ERA-interim in the troposphere (Jeuken et al., 1996; Jöckel et al., 2006; Jeuken et al., 1996; Löffler et al., 2016) towards the ERA5 meteorological reanalysis data (~~Berrisford et al., 2011~~) (Hersbach et al., 2020) of the European Centre for Medium-Range Weather Forecasts (ECMWF) to represent the actual day to day meteorology in the troposphere.

The setup of the chemistry submodels for this study is similar to the one presented by Jöckel et al. (2016, simulation RC1-aero-07), but with the addition of the submodel ORACLE (Tsimpidi et al., 2014) for the organic chemistry calculation and with stratospheric heterogeneous chemistry neglected. Initial conditions for the meteorology were also taken from the ERA-interim reanalysis data, while the ones for the chemical composition were from previous EMAC simulations (Pozzer et al., 2022). In addition, the anthropogenic emissions used are based on CAMS-GLOB-ANTv4.2 (Granier et al., 2019). To reproduce the effect of lockdown on the emissions, we adopted the reduction coefficient for Europe as in Guevara et al. (2021) for the sectors of energy production (ENE), road transport (TRO) and industrial processes (IND). The reduced emissions were averaged for the period 19th April to 26th of April (i.e. last available week in the dataset), and applied (for each country) for March, April, May and June. For aviation (AVI) we adopted the same method, although we applied the estimated factor to the entire aviation emissions, without any country distinction.

~~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~~ Atmospheric aerosols are described via a two-moment aerosol scheme, which predicts number concentration and mass mixing ratio ~~and mixing state.~~ Additionally, cloud formation 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) . Convective cloud processes are accounted for using the ~~convection~~ framework of Tost et al. (2006), based on the convection ~~calculation~~ schemes of Tiedtke (1989) and Nordeng (1994). ~~Large-scale cloud formations and prognostic variables depending on cloud microphysical processes follow the work of Lohmann et al. (2007); Lohmann and Hoose (2009); Baer et al. (2018)~~ . Thereby, the 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

165 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 , 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). 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).

Cloud droplet formation in the model setup without cloud-aerosol interaction is computed by the “unified activation framework”, an advanced physically based parameterization (Kumar et al., 2009; Karydis et al., 2011) that combines the κ -Köhler 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 ($T \leq 238.15$) $T < 238.15 K$), 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, autoconversion, 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.

The aerosol forcing of the EMAC model has been investigated, and here we report the Effective radiative forcing of the aerosol radiation interaction (ERF_{ari}) and the Effective radiative forcing of the aerosol cloud interaction (ERF_{aci}), based on the definition of Myhre et al. (2013). 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 $-0.46 \pm 0.01 W m^{-2}$ and $-1.2 \pm 0.1 W m^{-2}$ for ERF_{ari} and $ERF_{ari} + ERF_{aci}$, respectively. At BOA (bottom of the atmosphere) the model simulates $-1.6 \pm 0.02 W m^{-2}$ and $-2.1 \pm 0.1 W m^{-2}$ for ERF_{ari} and $ERF_{ari} + ERF_{aci}$, respectively.

We performed four simulations, all covering the period from January 2019 to July 2020:

- ~~STD~~ BASE : standard (i.e. "~~business as usual~~ baseline") emissions, without cloud–aerosol interaction,
- RED : reduced emissions due to lockdown, without cloud–aerosol interaction,
- 195 – ~~STD~~ CLOUD : like ~~STD~~ BASE CLOUD : like BASE but with aerosol–cloud interaction,
- REDCLOUD : like RED but with aerosol–cloud interaction.

In all simulations performed, the impact of different aerosol concentrations on the radiation (discussed in Sect. 4.2.1) is diagnosed but not used by the general circulation model, which instead adopts an aerosol climatology (Pringle et al., 2010). Similarly, changes in the tracers (e.g. ozone) do not influence the radiation, which is calculated with a greenhouse gases
200 climatology.

The model evaluation is performed with the RED simulation, while its difference with the ~~STD~~-BASE simulation is used to evaluate the impact of the reduced emissions during the lockdown. Simulation RED and ~~STD~~-BASE have binary identical dynamics (Deckert et al., 2011), i.e. they reproduce numerically exactly the same dynamics, as no feedback between chemistry and dynamic is present. Differently, in REDCLOUD and ~~STD~~CLOUD-BASECLOUD, the aerosol–cloud interaction is acti-
205 vated following the work of Lohmann and Hoose (2009); Bacer et al. (2018), leading to modification of cloud properties and therefore to changes in radiation and dynamics. The simulations REDCLOUD and ~~STD~~CLOUD-BASECLOUD are only used for estimating the indirect effects of aerosols (see Sect. 4.2.2).

2.2 BLUESKY observational data

We compare simulated trace gas and aerosol abundances to a comprehensive set of observations obtained during the BLUESKY
210 campaign (Voigt et al., 2021), ~~led by the German Aerospace Center (DLR) and the Max Planck Institute for Chemistry (MPIC), with the aim of investigating the effects of reduced emissions on atmospheric chemistry and physics. From 16th May to 9th June 2020 in-~~ In situ measurements of trace gases and trace particles were conducted end of May 2022 in the atmosphere over European urban areas and the North Atlantic flight corridor with the High Altitude and Long Range (HALO-) research aircraft and a second research aircraft, Falcon (see Fig. 1 for flight paths) Europe with the Falcon and HALO research aircraft. In total
215 8 and 12 flights were conducted with the HALO and the Falcon, respectively (Fig. 1).

We compare aerosol mass concentrations of black carbon (BC, size range between 70 and 500 nm), sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic aerosol particles (ORG, all from 40 to 800 nm) and aerosol particle number concentrations (between 250 nm to 40 μm). These are complemented by volume mixing ratios of carbon monoxide (CO), ozone (O_3), nitric oxide (NO), hydrogen peroxide (H_2O_2), peroxyacetyl nitrate (PAN), nitric acid (HNO_3), and sulfur dioxide (SO_2). Details
220 regarding instrumentation are provided by Voigt et al. (2021). We additionally use air temperature T , wind speed and specific humidity q to assess the quality of the reproduced synoptic conditions which are constrained (nudged) in the model. For the comparison, the model output was sampled ~~online in space and time during runtime~~ by the submodel S4D (Jöckel et al., 2016), following the flight tracks of the field campaign and with a time frequency of 5 minutes.

3 ~~Results:~~ Model evaluation

225 ~~A summary of the comparison of observations and model results is listed in and presented graphically in Figs. 2 and 3.~~

The ambient air temperature T is reproduced very well by the model; the average ratio of observed and simulated T is equal to 1.00 with a normalized root mean squared error of 0.04 (NRMSE; RMSE divided by range of observations). The vertical temperature profile is matched in the lower and free troposphere with a slight underestimation of observed temperatures towards

the upper troposphere (Fig. 2), which confirms the quality of the nudged data. Specific humidity q , a quantity that is not subject
 230 to nudging, is also captured reasonably well in the model, (NRMSE = 0.06), as 85.9 % of simulated values lie within a factor
 of two of the observations, yet slightly overestimated (see Fig. 2 and Table 1). In addition, horizontal wind speed $||\mathbf{u}_h||$ is also
 reproduced accurately with a low NRMSE (0.06) and an average ratio of 1.02.

Overall, the agreement between the meteorological variables from model and ~~observations~~ the one observed in the BLUESKY
campaign indicates successful initialization and nudging of meteorological variables and that the meteorological conditions
 235 during the relevant time period are simulated adequately. As the model is not nudged in the stratosphere or boundary layer
 (the nudging coefficient is maximal in the free troposphere (Jöckel et al., 2006)), the ~~slight underestimation of temperature~~
~~deviation~~ in the upper troposphere ~~region is not surprising~~ between model results and observational data are to be expected, due
to the intrinsic model dynamics which deviates from the nudging data. Nevertheless, the temperature bias is much lower than
 in other EMAC studies, despite the use of same nudging method and coefficients (Jöckel et al., 2016), due to the initialization
 240 and shorter simulation time in this work. As temperature and humidity are important quantities regarding cloud formation, and
 accurate wind vectors are key for representing advective processes, the following analyses of atmospheric composition and the
 effects on radiative transfer build on an accurate representation of the meteorological state of the model.

3.1 Trace gases

~~Observed~~ More than 94 % of simulated ozone (O_3) mixing ratios are ~~reproduced well by the model. More than 94 % of~~
 245 ~~simulated values are~~ within a factor of 2 of the observations ("PF2" value) ~~, and~~ the normalized root mean squared error of 0.04
 is low, with improvements from previous evaluation of the same model (Jöckel et al., 2016). Nevertheless, the model seems to
 slightly overestimate the observations, as already pointed out in various studies (e.g. Jöckel et al., 2016).

Simulated carbon monoxide (CO) mixing ratios are also in a good agreement with the observations, and virtually all simu-
 lated values lie within a factor of two of the observations. However, especially at lower altitudes, the simulated mixing ratios
 250 ~~somehow~~ underestimate the observed values, although the difference between average observations and average model results
 are well within their respective variability, and the shape of the vertical profile is qualitatively well reproduced. The same holds
 for nitric oxide (NO), which exhibits a C-shaped profile. The NRMSE for NO is low (0.08) and the average ratio of simulated
 to observed mixing ratio is 0.99, however more than a third of simulated values deviate more than a factor of two from the
 observations, due to the high variability of this tracer. Particularly the range of the observed mixing ratios close to the surface
 255 is not well reproduced by the model, which results from the short lifetime of NO and the challenge in reproducing its local
 variation by a global model.

Hydrogen peroxide (H_2O_2) and peroxyacetyl nitrate (PAN) are less well represented, the average ratios of simulated to
 observed mixing ratio (2.01 for H_2O_2 and 1.91 for PAN) indicate an overestimation by the model. Nevertheless, for both
 species about two thirds of the simulated points are still within a factor of two of the observations (see Fig. 2), and the
 260 measured dependence on altitude is captured by the model.

Sulfur dioxide (SO_2) was sampled predominantly at high altitudes between 370 to 170 hPa, where it is strongly under-
 estimated by the model. We hypothesize that the systematic underestimation of SO_2 concentrations is due to ~~an inaccurate~~

Table 1. Summary of model–observations comparison. The same spatio-temporal location were used for all simultaneously available points . NRMSE shows the root mean squared error normalized by the range of the observations. PF2 denotes the percentage of model points within a factor of 2 of the observations. The column $\overline{\text{MOD/OBS}}$ is the average of the simulated and observed data ratios.

Variable	NRMSE	PF2	$\overline{\text{MOD/OBS}}$
<i>Trace gases</i>			
O ₃	0.04	94.7	1.25
CO	0.14	99.3	0.98
NO	0.08	65.0	0.99
H ₂ O ₂	0.32	61.5	2.01
PAN	0.13	60.3	1.91
HNO ₃	0.37	12.9	0.46
SO ₂	0.40	25.9	0.43
<i>Aerosols</i>			
BC	0.09	18.6	0.68
NO ₃ [−]	0.14	20.6	0.92
NH ₄ ⁺	0.22	28.8	0.83
SO ₄ ^{2−}	0.16	26.8	0.72
Organics	0.45	40.6	1.73
Number conc.	0.11	42.8	2.60
<i>Meteorology</i>			
<i>T</i>	0.04	100.0	1.00
<i>q</i>	0.06	85.9	1.27
$ \mathbf{u}_h $	0.06	100.0	1.02

~~representation of transport from the boundary layer or from the stratosphere to the upper troposphere or due to~~ model shortcomings within the stratospheric aerosol chemistry, which will be discussed briefly as part of the following Sect. 3.2. All in
265 all, as summarized in Table 1, there is reasonable agreement between observed and simulated mixing ratios of the trace gases investigated.

3.2 Aerosols

In the comparison between model results and aerosol observations, the instrumental cut-offs have been taken into account; the aerosol log-normal modes in the model have been integrated only in the appropriate range, so to have a reasonable comparison.
270 In addition, all the measurements and model results are based on location pressure and temperature and are not normalized to Standard Temperature and Pressure (STP).

The vertical profile of the measured aerosol number concentration is qualitatively reproduced (see Fig. 3), with logarithmic scale in the x-axis, with a minimum at ≈ 300 hPa and a maximum at the surface. In the lowest altitude pressure bin, the range and median of the observations and model results match very well. There are some deviations between 850 and 480 hPa, where simulated number concentrations are larger than the observed ones, although this overestimation is well within the observations' variability. This overestimation dominates the average ratio of modeled to measured values (2.60, see Table 1).

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. The NRMSE of 0.09 is relatively low, as the higher abundance closer to the surface – that is, closer to the sources – is well represented, both in terms of magnitude and variability. A detail analysis of the black carbon concentration simulated with the EMAC model during the BLUESKY campaign can be found in Krüger et al. (2022).

Sulfate (SO_4^{2-}) exhibits qualitatively similar features as BC; the relatively high concentrations observed in the lower troposphere are matched by the simulated concentrations, yet there is a significant underestimation of sulfate aerosol concentrations in the upper troposphere. ~~We hypothesize that the modeled underestimation of sulfur species is related to missing contribution of volcanic eruptions that have reached the stratosphere at low latitudes and return to the troposphere at higher latitude. Many small and medium size eruptions have been reported in the year prior to the BLUESKY campaign (, last access 30. October 2021), but their influence on the upper troposphere and lower stratosphere is yet to be quantified. Some preliminary test simulations are mentioned below.~~

Between 1050 and 625 hPa simulated organic aerosol concentrations are somewhat larger in the model than in reality, the shape of the vertical profile is, however, qualitatively reproduced.

Nitrate (NO_3^-) and ammonium (NH_4^+) concentrations close to the surface are generally well reproduced. While, at higher altitudes, the simulated NH_4^+ agrees with the observations, simulated nitrate is too high, which is probably related to the co-located underestimation of sulfate.

The results of the model/measurement comparison are summarised in Table 1. We ~~conclude can observe~~ that there is generally reasonable agreement between simulated and observed trace gases and aerosols with some deviation of the aerosol concentrations, especially in the mid-upper troposphere.

A single ~~factor emissions source~~ 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 SO_4^{2-} do not correlate ($r < 0.01$, $p = 0.90$): in fact, mapping observed SO_4^{2-} concentrations to ozone (a tracer of stratospheric air), and carbon monoxide (a tracer of tropospheric air), reveals that high SO_4^{2-} concentrations coincide with high ozone ($r = 0.83$, $p < 0.01$) and low carbon monoxide ($r = -0.65$, $p < 0.01$) (Fig. 4). A similar, yet weaker, correspondence can be found in the simulated data (see Fig. 4). ~~This suggests~~ The strong correlation with ozone in the upper troposphere implies a stratospheric source of sulfate aerosols in both model and reality, ~~although stratospheric sulfate aerosol appears to be represented poorly in the model~~. It is noteworthy that a precursor for sulfate aerosols, sulfur dioxide, is also systematically underestimated. We assume hence that the high SO_2 abundance measured ~~is of volcanic origin~~ in the upper troposphere has stratospheric origin and is from volcanic eruptions. Many small and medium size eruptions have been reported in the year prior to the BLUESKY campaign (https://volcano.si.edu, last access 30. October 2021), but their influence on the upper troposphere and lower stratosphere is yet to be quantified. We tested

this by injecting high levels of SO_2 in the stratosphere in additional simulations, mimicking volcanic eruptions ~~at that had~~ enough energy to reach the stratosphere, i.e. Raikoke (June) and Ulawun (June and August) in 2019 (see de Leeuw et al., 2021; Kloss et al., 2021). However, this did not affect the concentrations of SO_4^{2-} and NO_3^- significantly (not shown). A partial
310 increase of SO_4^{2-} was obtained by including the volcanic eruption of Taal in January 2020. Nevertheless this is still not enough to bring the model results close to the observations. We therefore conclude that our observed underestimation of SO_4^{2-} is of stratospheric origin, although it is not fully clear what caused it.

A further partition of the region of interest into three subregions (Central Europe, Southern Europe, Atlantic) did not reveal substantial spatial dependencies of model deviation from observations (not shown).

315 4 ~~Results:~~ Impact of reduced emissions

To quantify the effect of the lockdown, we use the ~~business-as-usual simulations (STD and STDCLOUD)~~ baseline simulations (BASE and BASECLOUD) in the analyses. We focus on May 2020, as this time period is covered by the measurement campaign and the atmosphere can be expected to have adjusted to the impact of abruptly reduced emissions. We also analyse the impact in an area encompassing Europe (the region of study), i.e. over a longitude-latitude box from -20 to 20° E and 30
320 to 60° N (exactly the depicted map sector in Fig. 1).

4.1 Impact on tracers and aerosols

~~As no feedback between chemistry and dynamics is activated in the RED and STD simulations, any~~ As no difference in
dynamics between RED and BASE simulations are present, any chemical differences between these simulations are purely
attributable to the different emissions during the lockdown period, as these are the only changes between these two simulations,
325 and the consequent different chemical regimes.

In general, while large absolute changes are expected at the surface, in the upper troposphere (UT) we find the largest relative changes, due to the strong influence of the local emissions and to the low mixing ratios of most of the species investigated (see Fig. 5). Large relative changes in the UT are found for NO , SO_2 and BC , with a strong reduction ($\sim 50\%$ or more) in the region between 200 and 300 hPa, i.e. the typical aircraft cruise altitude. The reduced air traffic during the lockdown period
330 greatly decreased the emissions of nitrogen oxides into the UT, and the effects of the lockdown on other tracers in the UT are mostly a result of this strong reduction. Hydroxyl radicals (OH) decrease by roughly 20% in the UT and 5% elsewhere in the troposphere, a direct effect of a reduced OH recycling by NO_x . Despite the reduced OH , carbon monoxide does not increase, due to the decrease in the direct emissions. The overall effect of the lockdown for most ~~tracer-tracers~~ is a combination of reduced emissions and reduced sinks (i.e. oxidation via OH): while this is well balanced for CO (changes in the order of
335 few percent), for SO_2 the emission reductions are larger than the decrease in the reaction with OH , causing its mixing ratio to be reduced (up to 50% in the UT) compared to the ~~business-as-usual scenario. It must be stressed however, that those relative changes in the upper troposphere, although significant, have a very minor impact on most trace gas budgets, due to their low mixing ratios at these altitudes.~~ baseline scenario.

Similar to the trace gases, for most aerosols the lockdown reduces their concentration mostly at the surface, although the largest relative differences are simulated in the UT, due to the low concentration at these altitudes. For example, sulfate is subject to a large relative change in the UT but to much larger absolute changes close to the surface, mimicking the changes in SO_2 (see also Fig. 6). Furthermore, BC decreases significantly in the whole troposphere, due to the strong reduction of the emissions both at the surface and in the UT (from aircraft). ~~The aerosol reductions during the lockdown have implications on the incoming shortwave radiation, as discussed in the next section~~

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.

4.2 Impact on shortwave radiation

As the model is nudged in the troposphere (i.e. constrained air temperature with prescribed sea surface temperatures) ~~, we do not investigate any effects on the outgoing longwave radiation. Rather, we are free to adjust dynamics of the stratosphere, we report here RF (radiative forcing) values (Myhre et al., 2013). For the same reason (i.e. tropospheric nudging), we mostly focus the analyses on the shortwave flux F_{SW} and its induced heating rate $(\partial T / \partial t)_{\text{SW}}$ in the area encompassing Europe, as these are directly influenced by the aerosols changes and are not strongly influenced by the numerical forcing.~~

4.2.1 Direct effects

Aerosols directly impact the radiation balance by absorption and scattering of electromagnetic waves. Compared to the ~~business as usual baseline~~ emissions, the monthly mean sulfate (and inorganic aerosols, not shown) and black carbon concentrations are reduced ~~most strongly close to the surface, with another (much smaller) local maximum close to in all troposphere, with a strong relative reduction at~~ the commercial flight level (around 200 hPa, see ~~also~~ Fig. 6). Furthermore, the mean aerosol (number) concentrations were reduced in the ~~lockdown scenario scenario with reduced emissions due to lockdown~~ throughout the whole air column (see Fig. 8 and Sect. 4.2.2), with the reduction being most pronounced between 300 and 200 hPa, ~~likely. Based on model results from a sensitivity simulation, where only the aircraft emissions were reduced compared to the BASE simulation, we estimated that more than 90% of the reduced aerosols number between 300 and 200 hPa over Europe is due to reduced aircraft emissions and the effect on particle formation and coagulation at these altitudes.~~

We calculate the simulated difference in the downwelling shortwave flux between simulation RED and STD, i.e. the impact of the reduced emissions on the SW radiation. Here only the aerosol contribution is estimated, removing any radiative effect from changes in trace gases (e.g. ozone) within the Europe longitude-latitude box for May 2020. The differences are largest over continental central Europe and lowest over Northern Scandinavia (Fig. 7), with no large spatial gradients over Europe. In virtually all regions there is more downwelling shortwave radiation in the reduced emission scenario. Spatially averaged at ground level within the European domain, there is an increase of ~~0.327 ± 0.105~~ $0.33 \pm 0.10 \text{ Wm}^{-2}$ under clear sky condition (i.e., no clouds) compared to the baseline scenario, while at the TOA the increase is ~~0.198 ± 0.092~~ $0.20 \pm 0.09 \text{ Wm}^{-2}$. This increase, together with the reduced heating rates of ambient air, is indicative of a reduction in shortwave scattering and

absorption, due to the reduced inorganic aerosol and black carbon concentrations, i.e. the lockdown contributed to make the atmosphere more transparent to SW radiation. ~~We also compared our results with Van Heerwaarden et al. (2021): our radiative effect of all aerosols in our RED simulation for May is of -3.33 ± 1.36 , which is close to their value of -2.3 .~~ The column
 375 integrated contribution of backscatter and absorption can be estimated from the radiation values at TOA and surface, indicating that, during lockdown, the total backscatter (clear sky) of SW radiation has been decreased by ~~0.263 ± 0.070~~ 0.26 ± 0.07 Wm^{-2} , while the total absorption (clear sky) was decreased by ~~0.064 ± 0.053~~ 0.06 ± 0.05 Wm^{-2} . Based on an additional sensitivity simulations, in which only individual emissions (i.e. of BC, SO_2 , with NO) have been reduced, we found that slightly more than one third of this the absorption reduction is caused by the BC decrease.

380 Reduced scattering by aerosol particles plays a larger role, as the "net" (i.e. the difference attributable to the lockdown) shortwave flux is positive in the whole air column; on the other hand, reduced absorption dominates the shortwave component of direct aerosol effects in the boundary layer, as clearly shown in Fig. 6. The heating of ambient air exhibits a local minimum in the upper troposphere, which is however small compared to that in the lower troposphere. We calculate the surface integral of the accumulated heating due to shortwave fluxes, only attributable to aerosols under clear sky conditions: the difference
 385 in the atmospheric layer directly above the surface is -0.005 ± 0.001 K/day, i.e. less heating of the boundary layer in the lockdown conditions compared to normal emissions. ~~The decreased heating (for the entire column but mostly at the surface) is due~~ Based on a sensitivity simulation similar to RED but without any reduction in BC, we found that the decreased heating is by 40% to the reduced absorption by BC during the lockdown conditions, causing a cooling of the atmosphere (through SW radiation) despite an increase of the incoming radiation. Both the changes in heating and shortwave flux are solely attributable
 390 to the different aerosol burden in the ~~STD-BASE~~ and RED simulations.

We also estimated also the RF_{ari} Myhre et al. (2013) due to COVID-19 lockdown against the baseline scenario, by including also the longwave radiation. We obtained an RF_{ari} equal to 0.08 ± 0.03 for all sky over Europe in May 2020 at the TOA. This value, although accounting only for a limited amount of the anthropogenic aerosols (the lockdown did not removed all anthropogenic emissions) and referring only for Europe, is within the range suggested by Bellouin et al. (2020, see Tab.5).

395 4.2.2 Aerosol–cloud interactions

In Fig. 8, the vertical distributions of the total aerosol number concentration (N , including all aerosol sizes), ice crystal number concentration (ICNC), cloud droplet number concentration (CDNC) and ice crystal radius (r) are shown for Europe for both simulations, ~~STD-CLOUD-BASECLOUD~~ and REDCLOUD. Additionally, the SW flux at the TOA and the surface have been calculated from these coupled aerosol–cloud simulations (see Table 2), both for the total effect (i.e. direct plus indi-
 400 rect) and for the indirect (i.e. neglecting any direct radiation influence of the aerosol particles). Due to the short simulation period, the difference between these simulations is much smaller than its variability, represented by its spatial and temporal standard deviation. Nevertheless, comparing the vertical distribution of number concentrations of aerosols, ice crystals and cloud droplets, the largest relative difference between ~~STD-CLOUD-BASECLOUD~~ and REDCLOUD (i.e. the two simulations where the aerosol–cloud feedback is activated) is found for the aerosol number concentration between 200 and 300 hPa. These
 405 are the cruise altitudes at which the largest aircraft emissions are injected in the model and therefore these differences can be

directly connected to the reduced air traffic present during the lockdown (REDCLOUD). As this altitude is somewhat higher than the typical (cold) cloud altitude, the effect on clouds is less pronounced. At the highest level of these clouds (see Fig. 8) the ICNC are reduced (by $\simeq 30\%$ at 250 hPa, although with large variability), while no visible effect is found for CDNC. These results are in line with those obtained by Righi et al. (2021), who showed that aircraft emissions do increase ice crystal
 410 number concentration, although their results were not statistically significant. The ice crystal effective radius seems to be the least affected by the reduced emissions during the ~~COVID-19~~ COVID-19 lockdown, with a negligible absolute and relative difference.

To investigate the effect of reduced aircraft emissions on the SW flux via the indirect aerosol effect at the TOA and surface (SRF), the mean differences in SW flux between REDCLOUD and ~~STD~~CLOUD BASECLOUD for May were calculated
 415 over Europe. Positive values indicate greater reflection of SW radiation back to space (for TOA) or more absorption through the troposphere (for surface values) in the ~~STD~~CLOUD BASECLOUD simulation, compared to the REDCLOUD. The mean surface differences are 0.307 ± 0.115 0.31 ± 0.11 Wm^{-2} for the clear sky and 0.443 ± 1.063 0.44 ± 1.06 Wm^{-2} for the all sky case. At the TOA the mean differences in shortwave fluxes are 0.186 ± 0.106 0.19 ± 0.11 Wm^{-2} (clear sky) and 0.281 ± 0.928 0.28 ± 0.93 Wm^{-2} (all sky, Table 2). We should ~~notice~~ note that the clear sky results agree with the direct effect estimated in
 420 Sect.4.2.1 but with different simulations, confirming the consistency of the calculations. Thus, the indirect effect of aerosols enhances the direct effect on the SW radiation during the lockdown, even with larger intensity. ~~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)~~ –However, those values are associated with large standard deviations, related to the strong spatial variability of the upward shortwave radiation difference between the simulations.

The total RF_{aci} due to COVID-19 lockdown against the baseline scenario was also estimated. We obtained a value of 0.19 ± 0.92 for all sky over Europe in May 2020 at the TOA. Similarly to RF_{ari} , also this value is in line with the range suggested by Bellouin et al. (2020, see Tab.5), keeping in mind that only a partial reduction of anthropogenic aerosols took place during the COVID-19 lockdown. Although the average value agree with the literature, a large standard deviation is associated to RF_{aci} , so that the estimate should be taken with caution as not statistical significant.

430 5 Conclusions

We simulated the effects of drastically reduced anthropogenic emissions on the atmospheric composition in Europe during the ~~COVID-19~~ COVID-19 lockdown in spring 2020. We evaluated the model simulations with observations obtained during the aircraft measurement campaign BLUESKY. The overall agreement between observations and simulated aerosol concentrations and trace gas mixing ratios is reasonable. Nevertheless, problems remain regarding stratosphere–troposphere transport, especially of volcanic influence, which resulted in systematically underestimated SO_2 and SO_4^{2-} of stratospheric origin, and a consequent overestimation of NO_3^- ~~(which substitutes the underestimated sulfate in ammonium salts)~~ (which substitutes the underestimated sul
 435 in the upper troposphere.

Table 2. Aerosol direct and indirect effects on the shortwave radiation flux at the top of atmosphere (TOA) and surface (SRF) over Europe for May compared to baseline scenario. Note that direct effects are derived from ~~STD-BASE~~ and RED simulations, and indirect and total (i.e. direct plus indirect) effects from ~~STD-CLOUD-BASECLOUD~~ and REDCLOUD. The indirect effect of clear sky estimation is obviously equal to zero, but it was included to confirm the validity of the calculations.

$\Delta F_{\text{SW}} \text{ [Wm}^{-2}\text{]}$	RED- STD-BASE	REDCLOUD-BASECLOUD	
	direct	indirect	total
TOA	0.090 ± 0.035 <u>0.09 ± 0.03</u>	0.188 ± 0.759 <u>0.19 ± 0.76</u>	0.281 ± 0.928 <u>0.28 ± 0.93</u>
TOA clear sky	0.198 ± 0.092 <u>0.20 ± 0.09</u>	0.000 ± 0.006 <u>0.00 ± 0.01</u>	0.186 ± 0.106 <u>0.19 ± 0.11</u>
SRF	0.209 ± 0.053 <u>0.21 ± 0.05</u>	0.233 ± 1.089 <u>0.23 ± 1.09</u>	0.443 ± 1.063 <u>0.44 ± 1.06</u>
SRF clear sky	0.327 ± 0.105 <u>0.33 ± 0.10</u>	0.001 ± 0.023 <u>0.00 ± 0.02</u>	0.307 ± 0.115 <u>0.31 ± 0.11</u>

Focusing on the effects of aerosol particles on the shortwave radiation budget, we find that their reduction due to lockdown leads to a net clear sky SW flux increase of ~~0.327 ± 0.105~~ and 0.198 ± 0.092 0.33 ± 0.10 and 0.20 ± 0.09 Wm⁻² at surface level and TOA over Europe, respectively. The increase of the SW radiation during the lockdown period is due to the decrease of both black carbon and inorganic aerosols, which made the atmosphere more transparent to the incoming solar radiation by reducing SW absorption and SW backscatter, with the latter dominating. It must be stressed that although this BC reduction causes an increase in the SW incoming radiation, the SW heating has also been reduced by up to 0.005 K/day, due to the lowered BC absorption.

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 ~~effectively influence the cold cirrus clouds (aircraft cruising altitude, ≈ 250 hPa)~~ 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). 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".

Note that contrails and their contribution to radiative forcing are not considered in this study. Contrails are expected to reduce solar radiation reaching the Earth surface and to reduce outgoing longwave radiation. The mean changes induced by reduced air traffic in 2020 compared to 2019, computed in two model studies, were of the order of −0.1 to 0.5 Wm⁻² over Europe (Gettelman et al., 2021; Schumann et al., 2021b), ~~so at with~~ a magnitude comparable to ~~the aerosol effects~~ what found in this study. ~~We therefore plan to include contrail effects in a forthcoming study~~ The differences between these studies, can partly be attributed to the applied methodologies and general difficulties in discriminating anthropogenic effects from interannual variability. Hence, a study which considers contrail and aerosol effects simultaneously, and covers a longer time period, is recommended to better attribute the causes of the observed changes .

Code availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Website (<http://www.messy-interface.org>). The code presented here has been based on MESSy version 2.55 and is available as git commit #dc3ed8, in the MESSy repository.

Data availability. 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.

Author contributions. A.P. and S.R. planned the research. A.P. and S.R. collected and prepared the emission data. A.M. implemented code corrections for aerosol-cloud interactions. A.P. performed the model simulations. P.J. contributed to the overall model development and helped with the preparation of the model setups. M.K. provided the script for the aerosol mass estimation in the model. Z.H., I.T., L.R., D.J.C. and H.F. provided the data for CO, NO and H₂O₂. J.S. and K.K. provided observational aerosol composition data. R.D. and J.N.C. were responsible for the PAN measurements. C.V., L.T. and A.M. provided observational data of HNO₃ and SO₂. A.Z. provided the ozone data. O.K., B.H., C.P., M.P. and U.P. were conducting, analyzing and interpreting the BC data. M.D. organized the field campaign logistically. J.C. planned the flight tracks during the campaign. H.S. coordinated the measurements on the FALCON. S.R. and A.P. performed the model evaluation and analysis of direct effects. A.M. and A.P. performed the analysis of indirect effects. U.S. and A.P. discussed the results on the radiative forcing. S.R., A.M. and A.P. wrote the manuscript with the help of J.C., M.K. and J.W.. A.P. and J.L. supervised the project. All authors discussed the results and contributed to the review and editing of the manuscript.

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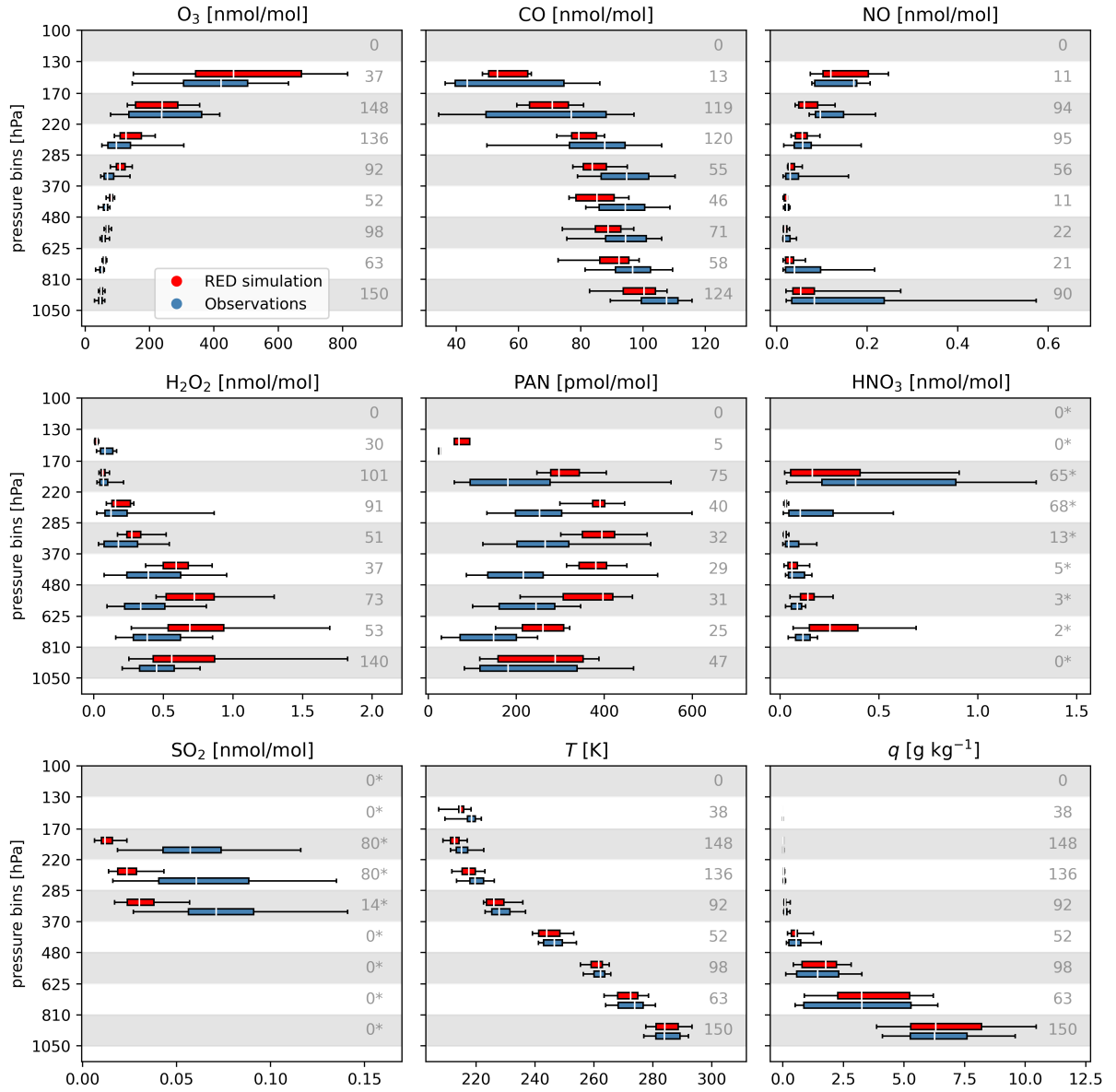


Figure 2. Vertical distribution of simulated (red, "~~MOD~~simulation RED") and observed (blue, "~~OBS~~") tracer mixing ratios and two meteorological variables (T and q), represented by box-whisker plots for pressure bins. The white line marks the median, the box corresponds to lower and upper quartiles, the whiskers represent the 5–95 percentile. The grey numbers on the right indicate the sample size (number of observed and interpolated simulated data points) for each pressure bin. Simulated values are from the RED simulation, i.e. with reduced emissions and no aerosol–cloud interactions. For HNO_3 and SO_2 (measured onboard of the Falcon aircraft, grey number marked with asterisks) the domain average of the model results over Europe at the corresponding altitude were used, not the values sampled online on the flight track.

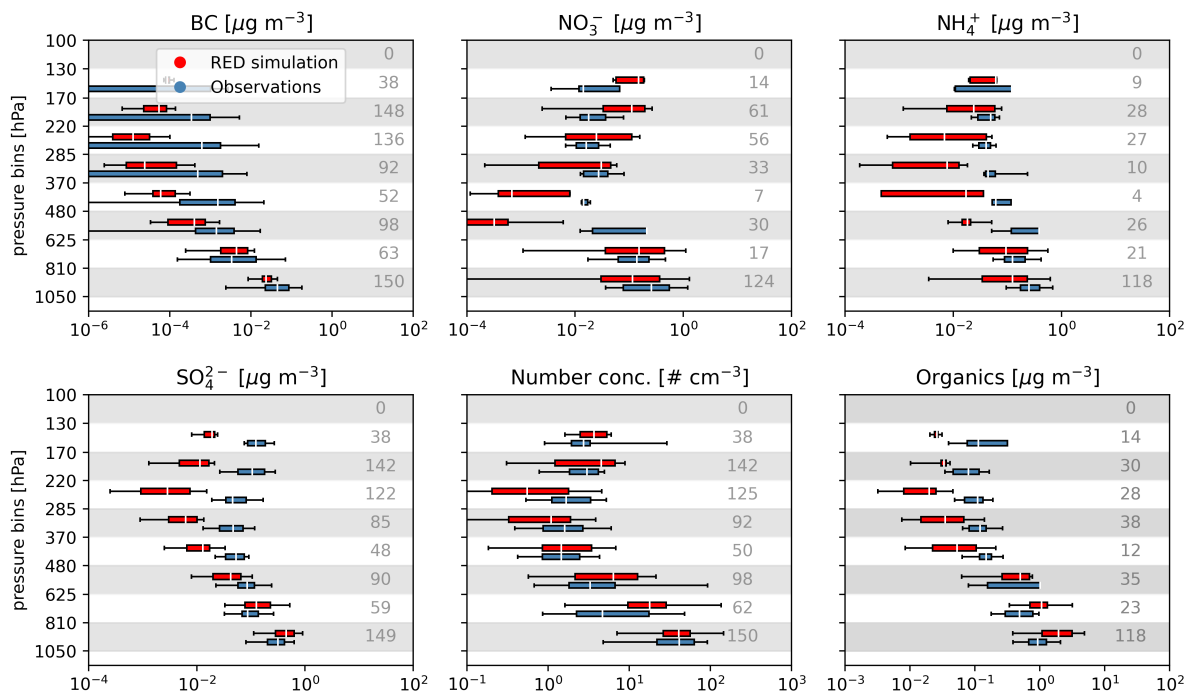


Figure 3. As Fig. 2, but for aerosols. Please note the logarithmic scale in the x-axis.

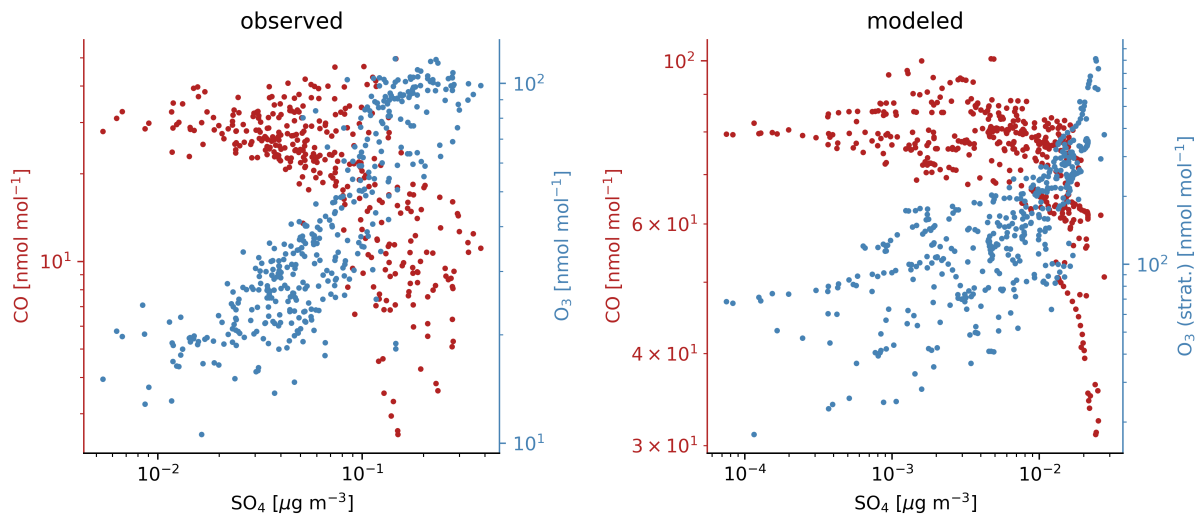


Figure 4. Scatter plot of co-located SO₄ and CO (red) and O₃ (blue) abundance between 350 and 150 hPa from observations (left) and the RED simulation (right).

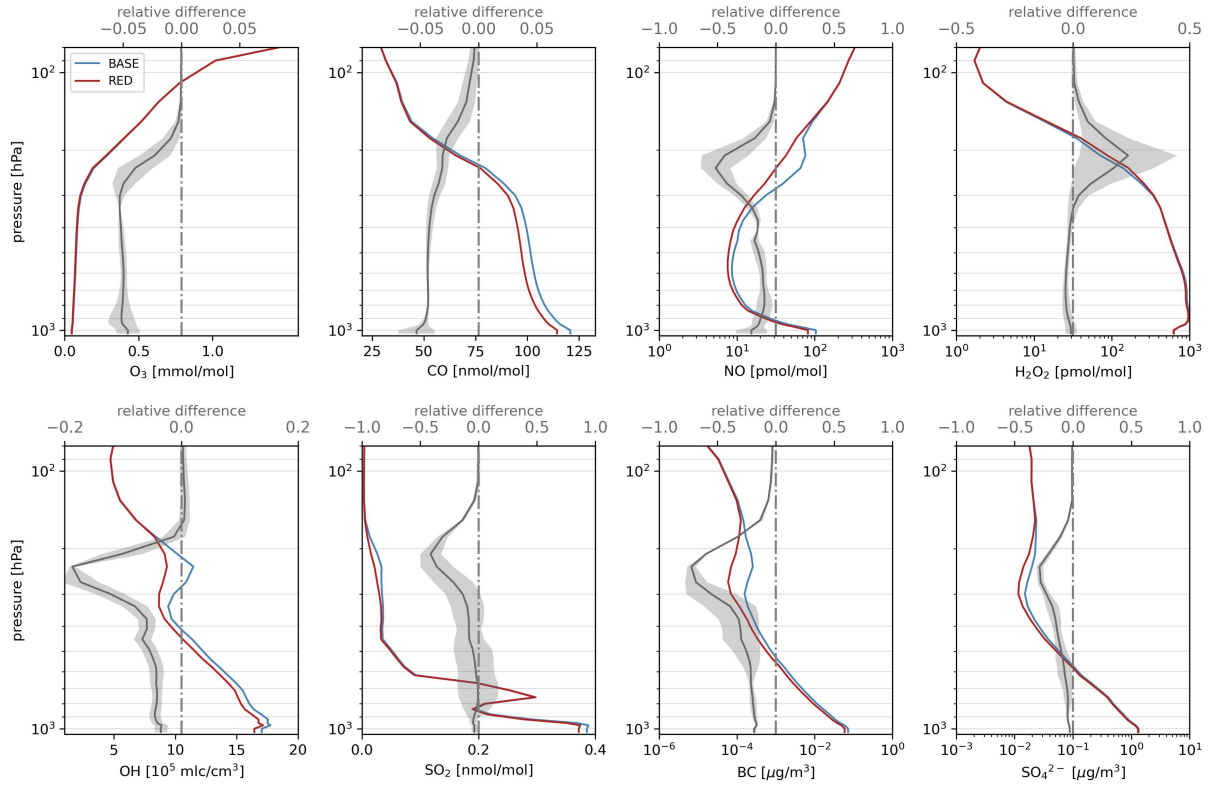


Figure 5. Vertical profiles from **STD-BASE** and RED simulations and their relative difference $((\text{RED} - \text{STD-BASE}) / \text{STD-BASE})$. The grey area represents one standard deviation of the spatial-temporal mean (grey line). Please note the different scales for the relative differences.

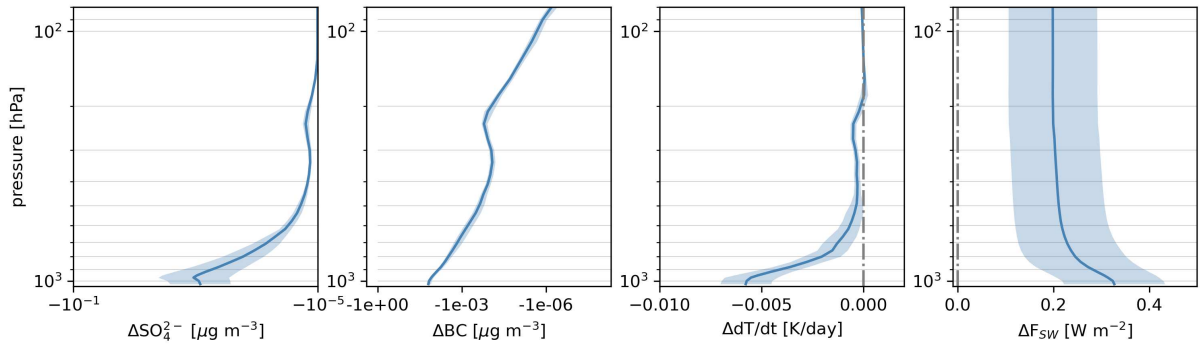


Figure 6. Vertical profiles of the difference of monthly mean sulfate mass concentration (SO_4^{2-}), black carbon mass concentration (BC), heating rate (dT/dt) and the net shortwave flux (F_{SW}) between the reduced emission scenario RED and the standard emission scenario **STD-BASE**. shortwave flux and shortwave heating are derived under clear sky conditions. The shading indicates one standard deviation of the monthly mean difference. Note the logarithmic horizontal axis for the two plots on the left.

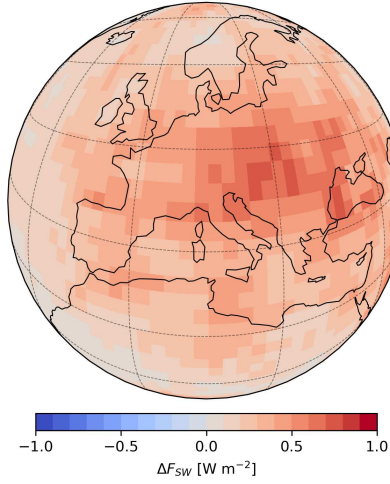


Figure 7. Difference of monthly mean clear sky shortwave radiation (May 2020) at the surface between RED and ~~STD~~-BASE simulation. Positive (red) values indicate more incoming radiation at the surface due to less absorption and backscattering in the "lockdown" atmosphere than in the ~~business-as-usual~~ baseline scenario. Note that we used a common reduction factor for emissions from countries outside Europe.

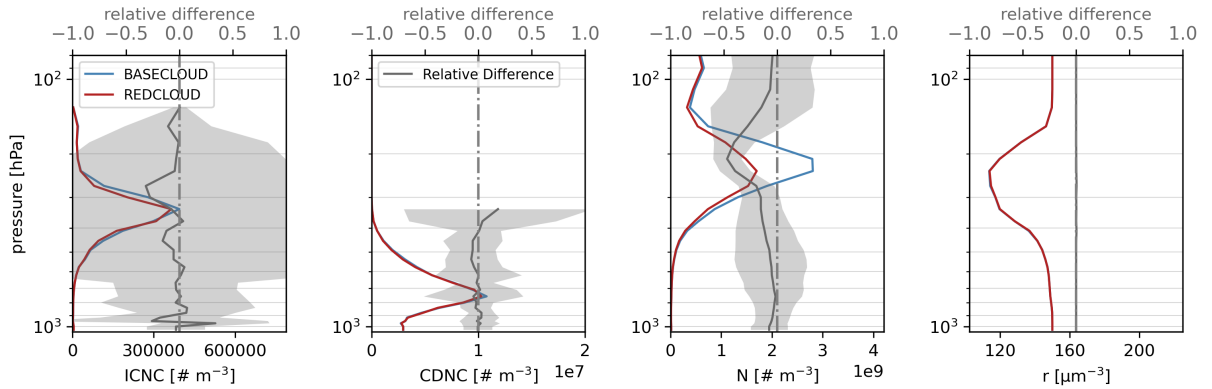


Figure 8. Vertical profiles of the monthly mean ice crystal number concentration (ICNC), cloud droplet number concentration (CDNC), aerosol number concentration (N) and ice crystal effective radius (r) of the reduced emission scenario REDCLOUD (red) and the standard emission scenario ~~STD~~-CLOUD-BASECLOUD (blue) and their relative difference (grey line) for May 2020 over Europe. The grey area denote the spatial and temporal standard deviation of the relative difference.