

We sincerely thank Referee#1 for the improvement suggestions. Below is a detailed answer to each comment which was raised. Changed made to the manuscript are reported in red hereafter.

Scientific Merit:

Significant changes in meteorology would be expected in the transition from winter to spring (i.e., pre-lockdown period and lockdown period) and even from early to late spring (i.e., beginning of lockdown period to end of lockdown period). Thus, the pre-lockdown and lockdown comparisons in pollutant concentrations (Fig. 1) and air mass trajectories (Fig. 2) are important for illustrating the point, but should not be a main focus for justifying the A3Q approach. For example, in lines 171-173 it is stated that the continental sector is under-represented and the oceanic sector over-represented when comparing the lockdown period with other reference periods. This is clear for the pre-lockdown period; however, within the measurement uncertainty there appears to be no statistically significant difference for the continental sector across the other reference years (over the same date range) and statistically different but (potentially) small differences for the oceanic sector in three of the four periods.

**The strong differences with pLP2020 actually flatten the differences with other reference periods. Still, we agree that this analysis may not be quantitative enough to prove the misrepresentation of the continental sector, although no measurement uncertainty is plotted in Figure 2 because no measurements were used in the trajectory analysis. We replaced it by a trajectory cluster analysis over LP2012-2020. The occurrence of continental air masses during LP2020 is around 28%, while it is 13-18% for the reference periods of previous years. The oceanic sector is inversely over-represented. The cluster analysis also reveals that highest NO<sub>3</sub> concentrations are associated to continental air masses (median of 6.6 µg/m<sup>3</sup>), which contrasts with the median value of 0.6 µg/m<sup>3</sup> for the oceanic sector. Therefore, an under-representation of the continental sector, as well as an over-representation of the oceanic sector, should lead to an underestimation of business as usual NO<sub>3</sub> concentrations during LP2020.**

Also following the comment of Reviewer 2, we changed section 3.1 as follows:

The assessment of lockdown impact on air quality lies on the use of a reference period, which is assumed to be representative of business-as-usual conditions during LP2020, following:

$$\%change = 100 \cdot \frac{LP_{2020} - ref}{ref} \quad \text{equation 1}$$

In the current literature, different “reference periods” are used, from a “pre-lockdown” period (pLP2020, Toscano and Murena, 2020; Dantas et al., 2020; Otmani et al., 2020), to the weeks corresponding to LP of previous years (e.g. 17/03 to 11/05 during 2017-2019 is LP2017-2019). Nevertheless, in the case of SIRTA, applying these methodologies unquestioningly, without verifying the inherent

hypothesis that data are comparable, can lead to significant variability, and counterintuitive results. Figure 1 presents concentration relative changes for the SIRTA dataset, using pLP2020, LP2019, LP2017-2019, LP2015-2019 and LP2012-2019 as references. Significant increases for all pollutants (e.g., + 83% in NO<sub>x</sub>, +439% in PM<sub>1</sub>) are found with pLP2020, which seems to contradict the observed drop of traffic. For the other reference periods, results reveal a substantial decrease for the concentrations of pollutants related to traffic emissions (i.e., BC<sub>ff</sub>, HOA and NO<sub>x</sub>), but clear increases of all other investigated pollutants, especially secondaries.

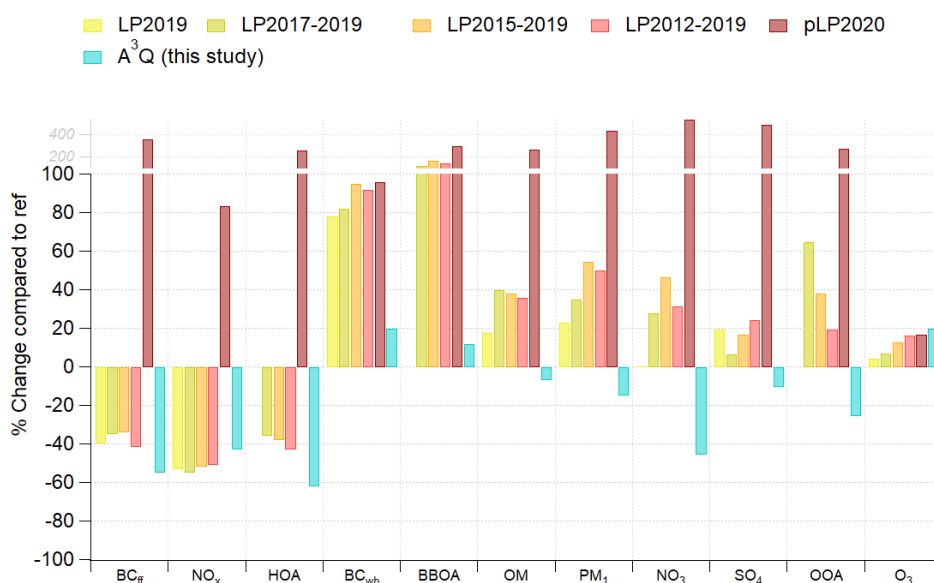


Figure 1 : Relative concentration change (%) of each specie used in this study following different reference periods, as well as from the A3Q approach presented in this article

As reference periods, they assume meteorological conditions representative of LP2020. However, April 2020 in France was exceptionally warmer (+4.5°C), drier (-43% of precipitation in the Paris region) and sunnier (i.e. hours of sunshine during the day; +60%) than usual (1981-2010 climatological reference values). Table S1 presents the meteorological variability of the different reference periods (in terms of ambient temperature, RH, pressure and wind speed), and shows that they don't reproduce the meteorology of LP2020 (in terms of min, max and average, especially T and RH), and also fail at reproducing its temporality (low r values). Moreover, from a trajectory cluster analysis (Fig. S3a), it appears that they misrepresent the variability of air mass origin. The unrealistic features of pLP2020 can indeed be explained by a drastic change of Western Europe meteorological conditions (from low-pressure to high-pressure system) concomitantly with the application of lockdown policy measures in France (Fig. 2). For the other reference periods, they still under-represent the continental sector (13-18%) compared to

LP2020 (28%), and inversely over-represent oceanic air masses. Given the fact that, for instance, NO<sub>3</sub>, SO<sub>4</sub> and OOA exhibit highest concentrations with continental air masses, these methodologies at SIRTA can most likely underestimate business-as-usual concentrations, and therefore lead to erroneous results.

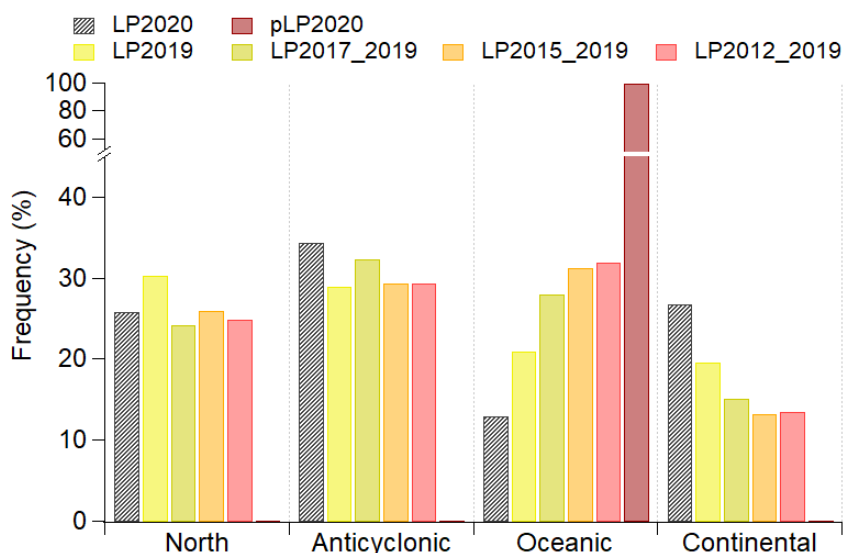


Figure 2. Frequency of trajectory clusters for each LP periods.

To overcome all these issues and account for the strong synergy between PM chemical composition, emission sources and meteorology, we developed the Analog Application for Air Quality (A<sup>3</sup>Q) method, which is described below.

Similar comparisons for temperature and precipitation for the LP and references periods would be valuable.

**A similar comparison for ambient temperature, RH, wind speed and pressure also highlights that these reference periods shall not be used in the case of SIRTA. We mentioned this in the text (see previous comment), and added a table in the supplementary:**

		LP2020	LP2019	LP2017-2019	LP2015-2019	LP2012-2019
Temperature (°C)	min	4.2	5.31	-0.5	-0.5	-0.5
	max	19.7	19.8	23.2	23.2	23.2
	mean	12.9	10.6	11.1	11.0	10.6
	MB		2.2	1.8	1.9	2.3
	r		0.17	0.4	0.45	0.45

RH (%)	min	34.0	36.9	36.9	36.9	34.9
	max	87.5	84.7	92.0	96.6	100
	mean	56.0	65.2	66.0	67.1	69.0
	MB		-9.1	-9.9	-11.0	-13.1
	r		0.36	0.32	0.28	0.30
Wind Speed (m/s)	min	0.8	0.3	0.3	0.3	0.3
	max	6.9	5.2	5.2	7.5	7.5
	mean	2.5	2.5	2.5	2.6	2.6
	MB		-0.03	-0.01	-0.13	-0.16
	r		-0.09	0.07	0.31	0.23
Pressure (hPa)	min	982.0	975.4	975.4	975.4	971.5
	max	1009.0	1012.5	1014.5	1014.7	1015.5
	mean	996.5	996.0	995.7	996.0	995.1
	MB		0.9	0.72	0.49	1.43
	r		0.13	0.03	0.08	0.05

Table S1: Meteorological conditions during LP2020, LP2019, LP2017-2019, 2015-2019 and 2012-2019. Min, max and average values for Temperature, RH, Wind speed and Pressure are presented for each period. MB and r are calculated through through a daily reconstruction of daily values of LP2020.

Minor comment-the date range listed in 161 does not match any of those in Fig. 1.

**As stated above, we changed section 3.1, but tried to make the date range clearer**

**“[...] to the weeks corresponding to LP of previous years (e.g. 17/03 to 11/05 during 2017-2019 is LP2017-2019) “**

The authors need to better demonstrate that the A3Q approach provides a significantly better solution than comparing with a range or ranges of previous years over the same days (i.e., see Parker et al. GRL, 2020).

**From the changes presented above, we think that it is now clear that meteorology needs to be taken into account, especially for secondary pollutants, whose concentrations at the receptor site highly depends on meteorological conditions. To this respect, lockdown periods of previous years can't be considered as representative of business-as-usual periods for the case of SIRTA. Then, we agree that the question “what is the performance of A3Q regarding meteorological parameters” would deserve a more detailed answer in the text. We changed this accordingly, with an additional figure in the supplementary:**

**“Despite these relatively wide ranges, the A<sup>3</sup>Q methodology allows to efficiently reconstruct meteorological conditions during the lockdown period. Indeed, Figure S11 presents, for the Jan.-May 2020 period, the temporal variations of observed and estimated Temperature, RH and Pressure. It shows low Mean Bias values, as well as satisfactory correlation coefficients (r value of 0.78, 0.82 and 0.63, respectively), which indicates a satisfactory analogy. Sensitivity tests presented**

below also demonstrate that stricter ranges do not significantly change the analog results. “

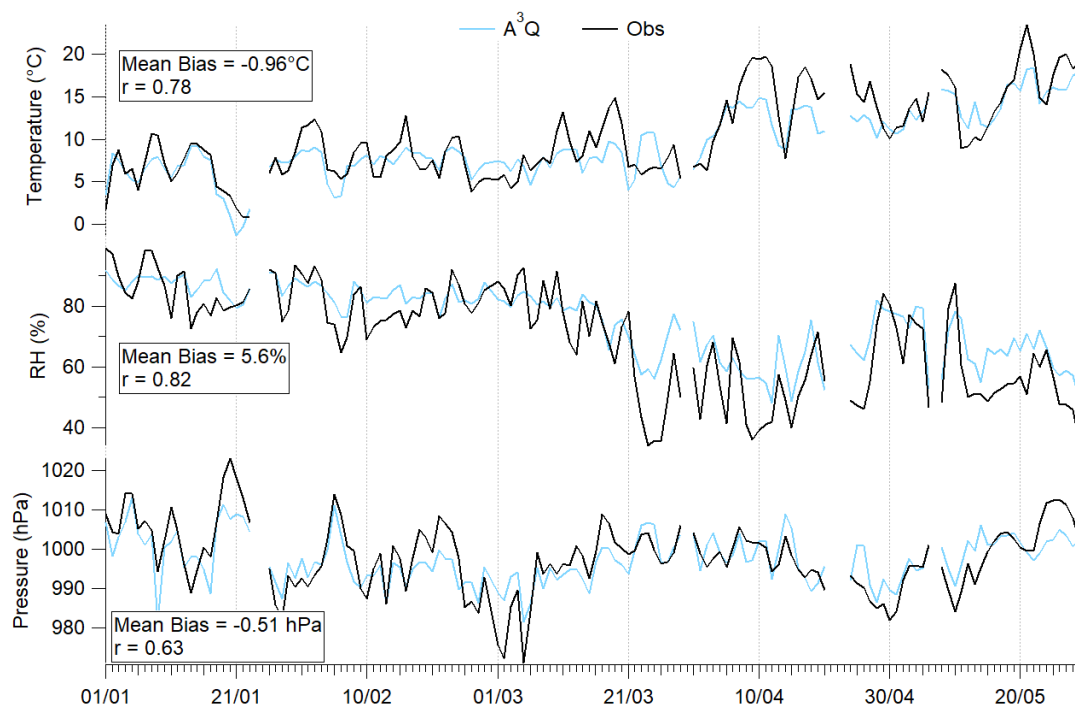


Figure S11. Temporal variations of ambient temperature, RH and pressure during January-May 2020, observed (black) and estimated by A<sup>3</sup>Q (blue)

It would be useful to see some of the pollutant concentrations presented in Fig. 1 with the results using the different analogs (Fig. 7).

**Figure 1 has been changed to add the results from A<sup>3</sup>Q.**

presentation quality:

Clarity of the manuscript could be improved with organizational changes and additional editing. The language in many places is quite challenging, and the main points are obscured or unclear.

The introduction, as written, includes one paragraph about COVID-19 restrictions, perturbations in human activity, and associated opportunities to better understand atmospheric composition and chemistry. Indeed, there have been a large number of studies published on air quality during COVID-19, some of which are cited by the authors. It is suggested that the introduction be reorganized such that these two pieces (motivation and current published studies) be combined as a single paragraph. The introduction could then be expanded to describe the current state-of-the-science regarding air quality in France/Europe and the importance of

considering meteorology in air quality studies, which are both relevant to the conclusions and only cursorily described in the second paragraph of the introduction. A more concise discussion of secondary chemistry and PM composition would also strengthen the introduction and the manuscript as a whole.

**Thank you for the suggestions. The introduction now reads as follows:**

“With the worldwide spreading of the SARS-COV-2 coronavirus, the COVID-19 outbreak has been responsible of millions of premature deaths. In order to slow down contagion rates, social interactions have progressively been limited until the establishment of strict lockdowns at national scales (Anderson et al., 2020) enforced during several weeks, especially during Spring 2020 in Europe. The corresponding stay-at-home orders resulted in a sudden halt of economic activities, and, as a consequence, in an unprecedented drop of emission of pollution sources. To this perspective, and despite tragic death records, these lockdowns are unique opportunities to characterize an extreme end of mitigation policy scenarios, and future low-carbon megacities from direct observations. Scientific initiatives are thriving across the globe in order to assess the impact of lockdowns on air quality. They report, for most, a sharp decrease of nitrogen oxides (NO<sub>x</sub>) concentrations, as well as an increase of tropospheric ozone (e.g. China: Le et al. (2020); India: Mahato et al. (2020); USA: Liu et al. (2020a); Europe: Sicard et al. (2020); Grange et al. (2020); South-America: Siciliano et al. (2020)) as a response to stay-at-home orders.

The increase of ozone is one counterintuitive example of the complex chemistry occurring within the atmosphere, although its link with the decrease of NO<sub>x</sub> concentrations has been well established (eg Reis et al., 2000). As highlighted by Kroll et al. (2020), beyond NO<sub>x</sub>, O<sub>3</sub> and PM<sub>x</sub>, additional information are needed in order to further characterize the impacts of lockdown on the atmospheric chemical system. Indeed, PM is composed of several different fractions, from organic to inorganic, and from primary to secondary pollutants, with diverse sources and transformation processes. Any concentration change of PM may derive from various compensatory feedbacks which are not characterized, limiting therefore our understanding of the impacts of lockdown on air quality. Moreover, Springtime in North-Western Europe is usually associated with high PM pollution episodes dominated by secondary material (mainly ammonium nitrate and sulfate, and secondary organic aerosols -SOA) as shown in Bressi et al. (2021). Ammonium nitrate is formed in the atmosphere from the neutralization of nitric acid (formed through NO<sub>x</sub> oxidation) with ammonia. The comprehensive characterization of SOA formation is also blurred by the overwhelming numbers of transformation pathways, precursors as well as oxidant availability. Thus far, only few studies have investigated the impacts of lockdown on PM chemistry and sources in Asia (eg Chang et al., 2020; Sun et al., 2020; Tian et al., 2021; Manchanda et al., 2021) by comparing the lockdown period with other periods (either a pre-lockdown period, or the same period of the year of previous years).

On the other end, the assessment of air quality implications of large cuts in urban pollutant emissions is strongly hampered by meteorological variability, which is one of the main drivers of air pollution temporality. For instance, unfavourable meteorology has previously been associated to increase of PM concentrations in various urban areas worldwide (eg Dupont et al., 2016; Wang et al., 2020; other ref). Sun et al. (2020) also highlighted severe hazes during lockdown in China, linked to stagnant meteorological conditions. Therefore, without climatologically representative values, specific care must be considered when comparing concentrations observed during and outside the lockdown period. The robustness of this assessment depends on the way meteorology is handled and on what “reference period” is chosen to compare with the “lockdown period”. A recent review by Gkatzelis et al. (2020) pointed out that, despite the luxuriance of scientific literature, more than half of examined articles didn’t take meteorology into account. Advances on machine-learning (ML) approaches have however enabled to disentangle the contributions of meteorological conditions on the temporal variations of primary and secondary PM components (e.g. Stirnberg et al., 2021). ML has successfully been applied mainly on NO<sub>x</sub> and O<sub>3</sub> in various European urban areas (Petetin et al., 2020; Grange et al., 2020). But weather-corrected studies of PM chemistry are still scarce, especially in Western-Europe.

The present study aims at reconciling a robust and innovative methodology with a quasi-comprehensive in-situ dataset, acquired within the Paris region (France). The 12-million inhabitants of the region, representing around 20% of the total French population, were placed under lockdown from March 17th, 2020 to May 10th, 2020, further designated as LP2020.“

There are several statements that are unclear as written. A few examples follow, but this is not an exhaustive list. It is recommended that the manuscript be reviewed for these unclear or ambiguous statements.

Lines 49-51: “Without climatologically representative values, comparisons of concentrations observed during and outside the lockdown periods shall thus free themselves from differences in weather.” The authors are making the valid point that care must be taken when comparing air quality measurements over different time periods because much of the variability can be driven by meteorology. However, this specific sentence seems to contradict that point, though it is very difficult to interpret.

**We rephrased the sentence as follows:**

“Therefore, without climatologically representative values, specific care must be considered when comparing concentrations observed during and outside the lockdown period“



Lines 55-58: The authors state- “Air quality shall not be restrained to NO<sub>x</sub>, O<sub>3</sub>, and PM<sub>x</sub> only, and limited number of studies so far has treated air quality as a whole, notably taking PM chemistry into account.” The sentences before and after suggest that the authors are referencing the need to consider speciation of PM. Depending on the objectives of a study, this may be a critical aspect, but isn’t necessarily a requirement for air quality studies.

**We agree. We rephrased the sentence as follows:**

“As highlighted by Kroll et al. (2020), beyond NO<sub>x</sub>, O<sub>3</sub> and PM<sub>x</sub>, additional information are needed in order to further characterize the impacts of lockdown on the complex atmospheric chemical system.”

Lines 176-177: “...it may also explain why these methodologies are associated to an increase of eg NO<sub>3</sub>, SO<sub>4</sub> and OOA, due to an underestimation of business-as-usual concentrations for LP2020 meteorological conditions.” It seems the authors here are suggesting that if the air masses for LP2020 were correctly represented/compared, and if there was no lockdown, then the NO<sub>3</sub>, SO<sub>4</sub>, and OAA concentrations in the reference periods may not have been higher than business-as-usual LP2020. However, there was no business-as-usual LP2020. Maybe the authors mean to suggest that the differences in concentrations, without accounting for meteorology/air mass origin, are not only due to changes in human activity and are likely exaggerated due to fewer air masses of continental origin in prior years.

**Yes, we definitely agree. Section 3.1 has been rewritten, providing further justifications.**

minor comments:

lines 79-83: The latter part of this sentence is unclear-it is clear that the measured fractions were corrected for collection and ionization efficiency, but it is not clear what it meant by “successfully participated in” intercomparison. Is this for the measurements being reported or is this something that has been done previously?

**We rephrased the sentence as follows:**

“and showed satisfactory performances during ACTRIS intercomparison exercises (Crenn et al., 2015; Freney et al., 2019).”

line 140: Please provide some additional details regarding ZeFir.

**We added additional details about ZeFir, as follows:**



“Calculations using HYSPLIT executables were automatically controlled by ZeFir (Petit et al., 2017a), a user-friendly interface based on Igor Pro 6.3 (Wavemetrics©). The cluster analysis presented in section 3.1 was also applied from HYSPLIT executables, controlled by ZeFir. Five clusters were used (Fig. S3a), in accordance with the Total Spatial Variance (TSV). The two oceanic cluster were summed as one.“

line 186: How is “sunny” quantified?

**It is quantified as the number of hours of sunshine during the day. We made it clearer in the text.**

“Additionally, April 2020 in France was exceptionally warmer (+4.5°C), drier (-43% of precipitation in the Paris region) and sunnier (i.e. hours of sunshine during the day; +60%) than usual (1981-2010 climatological reference values)“

line 227: Why would wetter days lead to enhanced condensation of semi-volatile compounds?

**Higher ammonium nitrate concentrations could be expected on days with higher RH, due to its hygroscopicity. Higher concentrations could also occur on colder days due to its semi-volatile properties. We corrected this in the text.**

“analogs that are much colder and wetter (higher RH) than the observation day may be associated to enhanced condensation of semi-volatile and/or hygroscopic compounds, which would lead to an overestimation of the estimated decrease of e.g. nitrate “

lines 230-231: Annotation (brackets) is unclear (also in Table 2).

**They mean “excluded”. We made it clearer in the text.**

“Acceptable ranges were therefore between the 5th and 95th percentiles (excluded) of  $\Delta T$  and  $\Delta RH$  values, which were respectively ]-9.3, 6[ and ]-19, 35[“

section 3.2.4-The presentation of the sensitivity tests is confusing as written, with both “S” used to indicate a scenario and also “scenario”.

**We replaced “S” with “scenario” throughout the section for consistency.**

line 295: “specie” should be “species”

**Changed.**