Reply to Zoltan Toth for the Editor Decision and Comments

First of all, we would like to thank the Editor for his comments and for giving us the opportunity to improve our paper. We added our acknowledgements to the editor in the new manuscript.

Now, we organized the answer to the comments as follows. First, we list some changes afford to the manuscript then detail our answers to the questions raised by the Editor in his decision and in the comments of Jan. 25th.

**List of changes for the revision**

***Minor changes***

- Fig. 2 and Fig. A1 'y' label has been modified using \chi in place of A.
- There was a typos in Eq.(D2) that has been corrected replacing \sum_{k=1}^N_e by \ sum_{k=1}^{N_e}

***Differences between the two versions of the manuscript***

To facilitate the comparison between the two version of the manuscript, a companion version of the manuscript lists all the modifications where old (new) statements are in red (blue). But the line numbers will refer to the revised version of the manuscript (not to the companion version).

**Answer to the comments**

We copied your commentary in italics below, we reply in normal blue font.

**I) Comment associated with the Editor decision:**

*1) The first and most important point is that both reviewers still find the manuscript excessively long (see Rev. 1's main comment, and Rev. 3's 1st comment). To address this, I encourage you to consider find Rev. 1's suggestion to move parts of the main text into Appendixes. Following her suggestion would shorten the main text significantly.*

We followed the recommendations proposed by Rev. 1 to move details in Appendixes, but kept a reduced version of the introduction of the univariate testbed at the end of the section on the background on the parametric formulation. The reason is that a large part of the material would have been moved in the multivariate part (details of the numerical setting e.g. time and space numerical scheme, illustration of the stationary wind used in Fig.1, description of the estimation of variance and anisotropy,..) and also because the description of the PKF dynamics helps to appreciate the effect of the chemistry in the multivariate framework (comparison between Fig. 2 and Fig. 6). See the detailed answer to Rev. 1

With these modifications, the manuscript has been significantly shorten.

*2) Another important point, first outlined in Rev. 3's 1st review (see 3rd par. in her/his first review) is further elaborated in the second review of Rev 3 (see her/his 2nd and 3rd comments in second review). [..] To address these points, you may consider corresponding changes wherever they are appropriate, including the Abstract, Introduction, main text, and/or Conclusions.*

*2.1) First, the subject is not specified clearly for your work - i.e., you may need to clearly state that uncertainties in chemical variables only due to chemistry, but not due to the dynamical evolution of the state are assessed in your work.*

This is now clearly stated :
- in the abstract “This contribution focuses on the situation where the uncertainty is due to the chemistry but not due to the uncertainty of the weather”(l 4-5)
- in the introduction “This contributions only focuses on the uncertainty dynamics due to the chemistry without accounting for the part of the uncertainty of the weather e.g. we do not take into account the uncertainty of the wind that transports the chemical species.” (l 92-93)
- in the conclusion “While a significant portion of the air quality uncertainty is due to meteorology (e.g. the uncertainty of the wind used for the transport), the present work focuses on the situation where the uncertainty in chemical variables is due solely to chemistry as it evolves along a given meteorological situation.” (l620-622)

*2.2) And second, the context and significance of the work need to be clarified accordingly - you may want to point out that in any practical application of the methodology you developed and tested, one must also address uncertainties in chemical variables due to uncertainties in the meteorological state variables. Introducing any ideas about how this could possibly be done is not a prerequisite of publication, but would certainly strengthen your report.*

The conclusion now addresses this point, and introduces how it would be interesting to take into account of the wind uncertainty in the framework of the parametric Kalman filter:
“In addition, since we have focused on the uncertainty due to chemistry, it would be interesting to address the part of the uncertainty due to meteorology. For a CTM like MOCAGE, this could be done by considering an ensemble of weather forecasts with each member used as a forcing for a single CTM forecast. However, this solution would lead to multiple CTM forecasts, which would be expensive. Therefore, from the perspective of using a PKF (applied to a CTM), a less expensive solution would be to consider a single PKF forecast where the wind is uncertain (stochastic advection wind), with the wind uncertainty characterized by the variance and anisotropy tensor estimated from the weather forecast ensemble. The challenge will be to find an appropriate closure for the unknown terms in the dynamics, including the cross-correlation between the wind error and chemical species, with the help of this contribution on multivariate statistics.”(l 654-661)

**II) Editorial comments on egusphere-2022-928, 25 Jan. 2023:**

*1) I found your responses to General comment 2 (G2), G2.1, Technical comment 1 (T1), and T6 from Rev. 1, where you state your preference to disregard reviewer suggestions lacking any reason or explanation. In your response to the next round of reviews, if you disregard any suggestion, can you please provide your reasons, beyond stating your preference? This is especially important in case of major recommendations, like the one highlighted in my next comment.*

Indeed, we should have explained better why we thought it was better to delete the parts that may have caused problems for Rev. 1 and Rev. 3. There were three reasons for this:

The first is that the PKF is new and that from experience it is not yet possible to rely solely on references -- even when they exist -- because readers want the contributions to be self-sufficient.
The second is that the multivariate framework chosen is complex since it takes into account both chemistry and transport. However, in order to understand which part of the uncertainty is linked to transport and which to chemistry, the passage through the univariate framework seemed to us unavoidable. Finally, we considered that the details provided contributed to the rigor of the work done, to avoid arbitrariness in the proposals we made.

At this stage, we did not think of moving what might have bothered the two referees to the appendix, as now proposed by Rev. 1. With this transfer to the appendix, the interested reader can follow the detail while making the manuscript much more readable.

*2) Rev. 1's G2 and Rev. 3's 1st general comments are aligned - they both criticize the excessive length of your manuscript. I sympathize with these comments. On the other hand, Rev 2 has not raised this as an issue. As we wait for the 2nd round of reviews, should Rev. 1 and 3 insist on this major recommendation, your options would include complying, at least partially, and/or explaining why keeping all material together would be beneficial.*

Following the recommendations of Rev1, the length of the manuscript has been significantly reduced now (as more explained in the answer to Editor decision point 1).

3*) Rev. 1, Specific comment 4 - wonder if the related additions are best placed in the Conclusions, or in Sections 3.3.2 and 3.5.2, where they may aid a more nuanced interpretation of the results?*

We introduced the nuance in the conclude section 3.3.2 (without change to the Conclusion where it is still mentioned): "It would be interesting to assess the robustness of the results, including whether the advection terms remain dominant under different conditions, such as weaker winds or accelerated chemistry, from a set of operational CTM predictions." (l 519-520)

4) Rev 1, Technical comment T2: is "autocorrelation" and "cross-correlation" the standard version of spelling?

We now use the terminology found in Derber and Bouttier 1999 who used "autocorrelation" and "cross-correlation", that is now better introduced in the manuscript "In multivariate covariance modeling applied in meteorology, these correlations are respectively denoted by autocorrelations and cross-correlations (Derber and Bouttier, 1999)." (l32-33)

5) Rev. 1, T7: what do you mean by your addition " prediction" related error? Do you want to refer to chaotically amplifying initial error in forecasts? If so, please clarify

We replaced "prediction" by "chaotically amplification of initial error in forecast" which is more clear now (see l 113)

6) Rev. 1, Technical correction TC38 - I find Rev. 1's suggestion reasonable. Much of Section 5 reads as a long summary. My understanding is that NPG has no strict rules as to the title of various sections.

We used the token \conclusion of the Latex template, that is now replaced by a section called "Summary and conclusions".

Reply to Annika Vogel – report 2

First of all, we would like to thank Annika Vogel for her review and for giving us the opportunity to improve our paper.

Now, we organized the answer to the comments as follows. First, we list some changes afford to the manuscript then detail our answers to the questions raised by the referee.

**List of changes for the revision**

***Minor changes***

- Fig. 2 and Fig. A1 'y' label has been modified using \chi in place of A.
- There was a typos in Eq.(D2) that has been corrected replacing \sum_{k=1}^N_e by \ sum_{k=1}^{N_e}

***Differences between the two versions of the manuscript***

To facilitate the comparison between the two version of the manuscript, a companion version of the manuscript lists all the modifications where old (new) statements are in red (blue). But the line numbers will refer to the revised version of the manuscript (not to the companion version).

**Answer to the question of the referees**

We copied your commentary in italics below, we reply in normal blue font.

*1. Main comment:*

*The total length of the manuscript was a point of discussion in the 1st review phase. I completely agree with the authors argumentation that each part of the manuscript is relevant. Indeed, all parts of the manuscript are important and contain interesting results. However, it's a combination of the actual length on paper and the amount of new content and results that makes the manuscript appearing very long. My main concern is that readers get discouraged or lost.*

*My recommendation is to move less important or "assisting" parts in the appendix - being still easily*

*accessible by the interested reader but reducing the length of the main text. I would suggest one or more of the following parts (or also others depending on the authors argumentation):*

*A) Sec 2.4 - univariate experiments: The univariate experiments in Sec.2.4 provide a good preparation for the subsequent parts especially for unexperienced readers, but the results are mainly an intermediate step for the interpretation of the multivariate results. Ideally this aspect could be addressed in a preceding publication, but could also be moved to the appendix of this paper. - this would shorten the main text by about 5 pages (in its current version)*

*B) Sec.3.2 - LV-CTM formulation: While the formulation of the PKF dynamics for the LV-CTM (Sec.3.2.1) and the multivariate PKF analysis (Sec.3.2.5) should stay in the main text, Sec.3.2.2*

*(evaluation of chemistry alone) and Sec.3.2.3 (contribution of individual terms) are interesting, yet less important parts which are evaluating the enKF results preparing the closure of the PKF equations. Thus, I recommend moving Sec.3.2.2 and 3.2.3 in the Appendix and referring to them in the actual formulation of the PKF closure in Sec.3.2.4 (e.g. adding reference to Sec.3.2.3 in l.555, ...).*

*- this would shorten the main text by about 5 pages (in it's current version)*

*C) Sec.3.3 - LV-CTM experiments: Algorithm 1 provides a good summary, but is rather long. It could also be moved to the appendix because all important steps are described in the manuscript. - this would shorten the main text by about 1 page (in its current version)*

*From my point of view, B) has more priority to be moved into the appendix than A) unless the authors argue differently.*

B) and C) have been moved entirely, and now correspond to Appendix D for B) and included in Appendix F for C) (algo 1).

For A) the description of the part related to the validation of the correlation functions (end of the former section 2.4.1) as well as the explanation of the significant difference between PKF and EnKF by the model error (former section 2.4.2) have been moved to follow the recommendations – (corresponding to Appendix A and B of the new version of the manuscript). However, we have chosen to keep the univariate example with the description of equations 9 and 10 (by deleting the title of sub-section 2.4.1) for the following reasons:

a) first the example introduces the numerical framework which should have been introduced anyway in the multivariate experiments, which would not have decreased the size of the paper, and this concerns the whole paragraphs "The numerical framework.." followed by the paragraph "For this experiment. " of l246-259). Indeed, these paragraphs introduce the details of the numerical resolution and the description of the wind field used (such as Fig.1).

b) then the example also introduces the diagnostics of ensemble estimation for mean, variance and anisotropy (paragraph "To assess..." l232-246)... which again should have been moved to the section on multivariate, so without impact on the size of the paper.

c) finally this example allows the understanding of the dynamics of uncertainties uniquely related to the conservation equation Eq.(9), which induces an evolution of the variance and span length fields (Fig. 2 of the new version), and thus prepares the reader for the interpretation of the multivariate framework (Fig. 6 of the new version) where one understands better what is the effect of chemistry in the propagation of uncertainties (visible by comparing Fig. 6(c) and Fig.2(b)). In particular, a clear reference to Fig. 2 has been added in results of Sec. 3.3.2.

Thus, compared to moving the whole section to an appendix, as you propose, and taking into account the displacement of paragraphs that this move would have induced (introduction of the numerical descriptions as mentioned in point a) and b) to be introduced in the multivariate part), the size of the paper corresponding to the introduction of the univariate part weighs only for approximately one and a half page in the current format (single column one row over two).

*2. Minor technical corrections:*

*a) related to the 1st review, reviewer1, technical correction 10: There are still some introducing sentences of next sections at end of sections. I suggest removing or moving them to beginning of new section (eg: before Sec.2.4, 2.4.1 ,3.1.3, 4.2)*

It has been at the beginning of Sec. 2.4 , 3.1.3, 4.2.

Section 2.4.1 title has been removed after the transformation Sec. 2.4 as discussed in main comment (move of point A) ).

*b) related to the 1st review, reviewer1, technical correction 9: Remove technical figure description "blue dashed lines" at l.314 and similarly l.419*

It has been removed now.

*c) l.43: Suggest reformulating the new sentence starting with "to zero" by moving these two words to the end: "...to set the ensemble estimation of the multivariate correlation to zero, …"*

It has been modified.

*d) l.194: In the current version the title of Sec.2.4 was not changed as described in the 1st review, reply to reviewer1, general correction 2.4*

Sorry, this is done now, thanks!

*e) l.106: Remove "that"*

We removed the word and added a comma before the word “and”.

*f) l.295-297: The complex sentence structure makes it difficult to follow. Do you want to say something like: Because the equation for the mean (Eq.(13)) is linear, the error field is given by an equivalent equation. And for these equation, the shorter the correlation length scale the larger is the error magnitude. ??*

We rephrase the paragraph as follows (in the new version where Eq.(13) now stands for Eq.(A1) ):

“This can be understood as follows. Since Eq. (A1) is linear, it is the dynamics of the mean and of the errors in the numerical experiment. But the typical scale of the mean and of the error are different: in this simulation, the spatial scale of the mean state is large, of the order of D, while the spatial scale of the errors is of order lh ≈ D/16, where 16 ≈ 241/15 ; this implies that the magnitude of the negative phase shift due to the dispersive term is larger for the error than for the mean (see e.g. KdV Eq. (1.19) in Whitham (1999), p.9).” (l694-698)

Reply to referee 3 – report 2

First of all, we would like to thank the referee for her/his review and for giving us the opportunity to improve our paper.

Now, we organized the answer to the comments as follows. First, we list some changes afford to the manuscript then detail our answers to the questions raised by the referee.

**List of changes for the revision**

***Minor changes***

- Fig. 2 and Fig. A1 'y' label has been modified using \chi in place of A.
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**Answer to the question of the referees**

We copied your commentary in italics below, we reply in normal blue font.

General comments:

*1. I suggested that the manuscript should be shortened because it tests patience of the reader with multiple examples that seem repetitious. That has not been addressed.*

Following the advice of Rev. 1 we moved a large part of the details in appendices, that shorten the new version of the manuscript.

*2. In my opinion the authors don't present "an application to a simplified chemical transport model" but present a concept of using PKF to propagating error covariances. There is big difference between application in a CTM model and illustrating a concept. Reference to CAMS ensemble that the authors mention in the reply is farfetched. In opinion it is hard to find any commonalities between CAMS ensemble and concept that authors present.*

In the new version of the manuscript we clarified the introduction of CAMS and the assimilation in MOCAGE :

We precise that CAMS is a multi-model ensemble ("..which daily forecast a *multi-model* ensemble of 11 members.." l 25), where each model has its own assimilation system ("Note that each member of the ensemble relies on its own data assimilation system for providing its surface analysis, while all models process the same set of surface observations, and all model forecasts are based on the same meteorological forcings from ECMWF high resolution weather forecasts." l26-29) and we precise that CAMS ensemble is not used within an EnKF ("In particular, members of the CAMS multi-model ensemble are not used within an EnKF to provide its own assimilation system." l29-30), all this to avoid any kind of confusion with the method and the framework we consider. Note also that we added a reference to the scientific description of CAMS (see confluence website reference in footnote 1, p2).

CAMS has been introduce to precise the state of the art, with a focus on the CTM MOCAGE and it 3DVar assimilation system that relies on simplifications we would like to avoid by using a PKF. This is now made more clear in the manuscript:

First we precise that the specification of the background error variance as a percentage of the first guess is very different from the forecast error variance that occurs in EnKF where it is deduced from the ensemble ("Note also that simplifications are often introduced to represent a flow dependency of the background term e.g. in several studies using MOCAGE, the 3DVar background error standard deviations are specified as a percentage of the first guess field (Amraoui et al., 2020; Aabaribaoune et al., 2021; Peiro et al., 2018) – which is very different from the forecast error variance in an EnKF that results from the ensemble estimation and the dynamics of the uncertainty." l39-43).

Then, we precise what would be the interest of using a PKF for the MOCAGE assimilation ("Compared to specifying the background variance as a percentage of the first guess, as mentioned above for the MOCAGE assimilation, the PKF could provide a flow dependence more consistent with the KF theoretical framework, but without the numerical cost of using an ensemble as with an EnKF." l82-85).

Since the MOCAGE assimilation and forecast system, as it is used in CAMS, is a 3DVar with background error variance specified as a percent of the guess and a forecast base on the deterministic high-resolution forecast of the ECMWF, we chose a testbed that reproduces a part of this configuration to tackle the multivariate framework: two chemical species in non-linear interaction, transported by a given heterogeneous wind over a 1D domain, with no uncertainty considered on the wind. Because of this it is a simplified CTM that share numerous important property of MOCAGE. This is now clearly stated in the manuscript with a clear reference to the MOCAGE framework: "To explore a multivariate formulation of the PKF, a simplified chemical transport model is introduced that mimics the MOCAGE framework. This simplified CTM contains the essential features of what can be found in a more realistic CTM, that is advection, multiple chemical species and non-linearities." (l298-300).

*3. Reply to my 3rd inquiry is long but, in my opinion, entirely misses the point. Whether the model has interactive meteorology and chemistry has no relevance to the data assimilation approach discussed in the manuscript. In a common application the spread of an ensemble of chemical model realizations may come from varying meteorological states, be it wind in the simplest case for off-line model (u,v) plus state variables (T,q,...) in an online model. Varying meteorology will contribute to spread of the chemical ensemble because it will affect concentrations of the species. As noted in my review, I don't believe that the approach presented will lead to an efficient data assimilation. To be convinced I would welcome an application that the authors proposed using MOCAGE. Otherwise, as I pointed in my review a theoretical basis for*

*a concept may exist but the concept itself will remain just a curiosity without any prospect for a real-world application.*

As detailed in the answer of point 2), the 3DVar assimilation of MOCAGE (as used in the CAMS operational ensemble where MOCAGE is one of the state of the art model), do not take into account the variability due to the meteorology. We want to improve the assimilation of MOCAGE using a PKF, while keeping a low numerical cost for the improvement. However this contribution focuses on the multivariate issue, and the uncertainty due to the chemistry as it is now clearly stated in the abstract (l 4-5), in the introduction (l92-93) and in the conclusion (l620-622) (see our detailed answer to the Editor decision point 2.1) which is about this focus).

Note that the specification of the background variance in the 3DVar of MOCAGE is a way to take into account a part of the uncertainty due to the meteorology (since it is flow dependent through the first guess), but this is far from the uncertainty due to the meteorology as it would follows from the considering the Kalman framework that would be based on en ensemble of weather forecast. After considering the multivariate issue, we are agree that introducing the uncertainty due to the meteorology is an important issue. However, this point can not be addressed without any clue on how to consider the multivariate statistics e.g. the cross-correlation of between the uncertainty on the wind and on the chemistry. The present work contributes to gives some ideas about how to do that.

In particular, in the conclusion, we indicate how we think interesting to take into account the spread due to the meteorology in a way consistent with the PKF approach:
“In addition, since we have focused on the uncertainty due to chemistry, it would be interesting to address the part of the uncertainty due to meteorology. For a CTM like MOCAGE, this could be done by considering an ensemble of weather forecasts with each member used as a forcing for a single CTM forecast. However, this solution would lead to multiple CTM forecasts, which would be expensive. Therefore, from the perspective of using a PKF (applied to a CTM), a less expensive solution would be to consider a single PKF forecast where the wind is uncertain (stochastic advection wind), with the wind uncertainty characterized by the variance and anisotropy tensor estimated from the weather forecast ensemble. The challenge will be to find an appropriate closure for the unknown terms in the dynamics, including the cross-correlation between the wind error and chemical species, with the help of this contribution on multivariate statistics.”(l 654-661)