We thank the reviewers for taking time to provide helpful comments, recommendations and insightful questions that helped the author to improve the paper.

In the following, the issues and remarks of the reviewers are individually addressed unless they were simple typographical or technical corrections, which we simply applied. Comments of the reviewers are repeated for convenience as indented blocks. Within the provided replies, excerpts from the revised paper are marked by cursive face.

1 Reply to Referee 1

1.1 Major Remarks

1. > Detailed presentation of mathematical methodology -duplication with previous work
   >
   > There is quite some redundancy / repetition in the presentation of the retrieval approach - Gauss-Newton iteration, setup of regularization matrix, averaging kernel and gain matrix diagnostics, etc. . . . - when comparing this manuscript with the previous papers mentioned above, nb. the two GLORIA papers. For the reader of this manuscript (including the reviewers) it is therefore cumbersome to identify the advances presented in the current manuscript. A detailed presentation of the methodology has been clearly useful in former times, when papers were difficult (or sometimes almost impossible) to obtain. However, nowadays literature is mostly accessible by just a few mouse clicks". In particular, all previous Ungermann et al. papers have been published in an Open Access journal (AMT(D)) and therefore are readily available.
   >
   > As a consequence I suggest to rewrite the methodology section 2 in a significantly more compact manner, clearly indicating the differences to the previous presentation.

   I agree that there is some redundancy with past papers. It is however felt that most of this repetition is necessary for two reasons: first, the interested reader should not be forced to look into the other papers to understand the current one (while she is certainly invited to); second, presentation of technical details for deriving a full set of diagnostics is helped by introducing and explaining the preceding formulae.

   Still there is indeed information that is unnecessary for the purpose of this paper that may be removed. The paragraph referring to Tikhonv/optimal estimation was removed (6582 l.10-18) and the pragraph describing the employed regularisation (6583 l.17- 6584 l.10) was shortened to "*As compromise, the Tikhonov regularisation used in this paper is chosen to approximate the precision matrix of an optimal estimation covariance matrix employing the auto-regressive model to fill the covariances (e.g. Steck and von Clarmann, 2001). The parametrisation used here follows closely the one described by Ungermann et al. (2012) with the notable exception of the added* *matrix for horizontal regularisation. To summarise the setup briefly,* $\mathbf{L}_0 \in \mathbb{R}^{n\times n}$ *is a diagonal matrix, with climatological standard deviations on the diagonal. The* $\mathbf{L}_1$ *matrices pose constraints on the first-order derivative in vertical and horizontal direction, scaled with two quantity specific scaling factors* $c_{\mathrm{q}}^{\mathrm{h}}$ *and* $c_{\mathrm{q}}^{\mathrm{v}}$ *for* $\mathbf{L}_1^{\mathrm{h}}$ *and* $\mathbf{L}_1^{\mathrm{v}}$*, respectively.*" Some cites were also added to help the reader identify repetitions of previously published material; e.g. the "Linearised diagnostics" section is introduced by the more verbose "*The diagnostics used in this work follow the linearised diagnostics described by Rodgers (2000). The key point of this section is how these well-known diagnostics may be derived in a memory conserving and numerically stable way required for large-scale retrievals. It thereby expands the previous work only detailing the calculation of the noise error (Ungermann et al., 2010) to the more complicated estimation of systematic errors induced by background gases and uncertainties in spectral line data.*"

2. Noise in retrieved state vector

   The term noise used frequently in this manuscript to describe some properties of the retrieved profile(s) is not appropriate. Clearly the measurement (vector) is contaminated by noise, however, the retrieved profile(s) (or their discretized representation, the state vector) can have instabilities, oscillations, . . . in case of insufficient regularization.

The author does not fully agree with the reviewer comment. Clearly, an ill-posed problem such as discussed here can introduce strong oscillations and other artefacts by magnifying noise during the retrieval. But due to proper regularisation, the discussed case study should be free from such artefacts. Still, even if the problem were well-posed, instrument noise will reflect on the retrieved VMRs. Gaussian noise or spikes in the measurements cause in a first order approximation similar structures in the retrieved profiles. Plotting cross-sections of measured radiances and comparing them with retrieved trace gasses shows the striking resemblance of structures including obvious artefacts. To some extent, the effect of instrument noise (Gaussian and spikes) is reduced by the smoothing properties of first-order regularisation. Also, as the cross-section retrieval produces images, terminology common to describe artefacts in (photographic) images seems appropriate here.

To distinguish this notion, the paper now uses the term "image noise" when referring to visible noise and delivers the following definition of the term: "*Image noise refers here to the artefacts induced by measurement noise and similar stochastic errors in the radiances.*". The image noise is already quantified in the noise error and referred by that when discussed quantitatively. Further, where appropriate the terms "measurement noise" and "noise error" are used instead of the ambiguous "noise".

1.2 Minor Remarks

1. 6580.14 "This is typically accomplished by adding constraints . . . " Are there other ways of regularization?

Other regularisation methods are the (also employed) discretisation of the underlying continuous problem, the early stopping of iterative algorithms used for solving involved (linear) equation systems, or using a singular value decomposition to identify and discard small eigenvalues of the linear equation system matrix. These methods deliver robust results but are not able to fully exploit the a priori knowledge available for atmospheric retrieval problems. But they are popular in other fields.

2. 6581.04 The second sentence is incorrect, not only limb sounding inversion is illposed, nadir sounding is even worse (as correctly stated in the final section).

The given sentence does not try to make any statement with respect to the ill- or well-posedness of retrieving from nadir sounder measurements. It talks only about limb-sounders. The noted sentence is further redundant with the introduction and was removed in the revised version. The introduction is accordingly modified to: "*The retrieval of trace gases or other quantities from infrared nadir- or limb-sounder measurements is inherently an ill-posed problem, . . .*".

3. 6581.06 ". . . representation of the atmospheric state x is modified . . . until the fit . . . is deemed good enough . . . " This sounds like an iterative procedure, which is clearly required for nonlinear problems. However, linear (small-scale) problems can be solved in just one step without iteration.

This is indeed the case. The author sees linear problems as a sub-case of non-linear ones, as they can be treated with the same methods as the non-linear ones. In case of linearity, the iterative solver will terminate after the first iteration if proper stopping rules are in place. The following sentence should help the dissenting reader: "*If the forward model is linear, a solution can be directly calculated while non-linear forward models require an iterative procedure.*"

4. 6583.19 ". . . insert the minimum of the cost function xf . . . " Rephrase! xf is not the minimum of the cost function, rather it is the x minimizing the cost function.

This and other misuses of minimum were addressed.

5. 6590.18 ". . . get reduced in lockstep . . . " — Please explain

If the standard deviation of the error estimate is reduced, so is the frequency of large rel. errors in the estimate. The sentence seems to be confusing as the content should be self-evident. It was removed from the paper.

6. 6591.11 "This data set is rather unique . . . " — Does this refer to the entire campaign data set or just the second flight on 2. March??? If necessary move this sentence down or the very last sentence up.

It is clarified as "*One of the instruments aboard was CRISTA-NF, an airborne infrared limb-sounder. The data taken by CRISTA-NF in this campaign is rather unique in having at the same time a high frequency of taken profiles (one profile every* $\approx$ *15* km*) and a high vertical sampling (*$\approx$ *250* m*). . . .*"

7. 6591.23 ". . . from the flight altitude down to 15 km below . . . " — This is quite confusing. According to Ungermann et al. [2012] the scan goes down to 5 km.

The sentence fails at communicating that the vertical coverage of the instrument is 15 km. It is remedied by a simpler "*Spectra are scanned from the flight altitude down to* $\approx$*5* km *in vertical steps of* $\approx$ *250* m *using a Herschel telescope with a tiltable mirror.*"

8. 6592.24 Retrieval setup: it would be helpful to indicate the (total) length of the measurement vector and of the state vector.

The sentence "*In total, this gives a state vector* $\vec{x}$ *with 93 870 entries and a measurement vector* $\vec{y}$ *with 73 660 entries.*" was added.

9. 6593.04 "All targets are derived between 0 km and 25 km . . . " Probably the lower limit is essentially the lowest tangent height!?!

The lower limit for "useful" values is mostly defined by the lowest tangent height. Though sometimes, the signal to noise ratio further restricts the usefulness of derived values as it happens for $ClONO_2$. Still, trace gas VMRs are being retrieved for all altitude levels down to 0 km, whereas the lowest ones are obviously fully determined by a priori information. For 1-D retrievals the difference in computational effort between a lowest limit of 0 km or, e.g. 5 km is completely negligible. Having a large safety margin prevents problems stemming from limb-rays passing unexpectedly below the lowest retrieval limit. For cross-section retrievals, the wasted computational effort is noticeable but still not worrisome. Adding or removing the lowest altitudes does not meaningfully affect the time required to calculate the Jacobian matrices. The time to solve the linear equation system *is* affected by the superfluous entries, but this time is dominated by a factor of three to ten by the time to compute the Jacobians. However, it is planned to use a more sophisticated choice for the lower bound depending on the lowest measurement for cross-section and tomographic retrievals in the future.

10. According to Ungermann et al. [2012] the retrieval grid above 30km has a spacing of 2 km?

The text was corrected to "*The retrieval grid sampling distance is 250* m *below 20* km*, 1* km *between 20* km *and 30* km*, and 2* km *above.*"

11. 6596.18 HITRAN11: the HITRAN 2008 database [Rothman et al., 2009] including recent updates?

The reviewer assumed correctly. A reference to Rothman et al. (2009) was added and the “11” was removed.

12. 6600.25 ”. . . being the retrieval being nonlinear).” — Isn’t the retrieval nonlinear anyway???

The following should express the intended meaning clearer: “*This corresponds to a maximum likelihood estimator and is mathematically similar to a linear cross-section retrieval linearised at the state given by the assembly of the 1-D solutions.*”

1.3 Technical Remarks (typos etc.)

1. 6584.13 Move opening parenthesis to front of citation

I do not see how to apply this comment. The given style of cite “by Rodgers (2000).” is consistent with the AMT style guides with respect to citing.

2. Figs. 2, 3, 4, 5, 11 Title of the plots ”retrieval results” ???

The rather redundant titles of “retrieval result” and “horizontal resolution” were replaced with titles describing the regularisation strength of the depicted retrieval.

2 Reply to Referee 2

2.1 Specific Comments

1. Page 2 line 25: Points out that filaments of a lesser extent than the measured vertical resolution are not resolvable, but in the context implies that this algorithm could do so. Surely if the measurement density is lower than the atmospheric feature then that information is lost?

The paper notes that the vertical resolution shall be improved compared to conventional techniques. In either case the lower limit for the vertical resolution is given by the vertical sampling or the measurement density. Due to regularisation, the vertical resolution is often noticeably worse than the vertical sampling or the measurement noise makes it difficult to identify faint structures. To clarify this, the sentence “*Obviously, the lower limit for the improvement is given by the vertical sampling, which needs to be fine enough to sample structures of interest.*” was added.

2. Page 9 line 16: Here a part of the elaboration of the algorithm is omitted due to the complexity of the notation. In the other parts of the manuscript the author is very explicit in describing the mathematics of the algorithm. A choice should be made if a full formalism is desired to allow third parties to recreate every single step of the work, or if only novel steps need to be

described. In the first case the missing steps should be included; in the second case the author could simplify the whole of section 2 and refer to the literature for well established aspects of the optimal estimation algorithm.

A mixed approach was followed to address this comment. Material repeated from previous publications was reduced to the necessary and text was added that helps the reader to identify novel material (see Major Remark 1 of Reviewer #1 above). In addition, the missing details of the algorithms were supplanted in an appendix for completeness' sake.

3. Page 13 line 9: Would this study have worked for another flight date as well? Is some date better suited than others? If yes which ones, and why.

The figure is representative for any kind of study. I added the sentence "*The quality of the estimates follows the expected theoretical forecast, which makes a separate calculation superfluous.*" to highlight the connection between the generally valid theoretical forecast and the practical example.

4. Page 14 line 6: A tangent point uncertainty of 100m is calculated from a given pointing angle uncertainty, but due to the limb-viewing geometry this number will vary as a function of the current scan angle. What is the absolute tangent altitude at which a 0.02 deg pointing error results in a 100m tangent point error?

The value was given as a rough estimate. This estimate was restated more precisely as "*which corresponds to an uncertainty in the tangent point altitude of* ≈ 125 m *vertically 10* km *below flight level.*".

5. Page 15 line 20 (also, page 20 line 23): "The choice of CFC-11 is disadvantageous for cross-section retrievals." Why chose this gas then in the first place?

To demonstrate a positive effect even under adverse circumstances. Any new technique is beneficial for carefully controlled and selected use cases. We make a point by using our most recently published data set and select from that the least and most benefiting trace gases.

Further, having two data points (one gas with a strong signature and one gas with a weak signature) allows thereby (to a certain extent) to extrapolate the expected gains when applying the presented technique to other instruments.

We added "*Thus, the two trace gases represent a worst case and a best case scenario for the proposed algorithm and thereby allow a better quantification of expected benefits for other scenarios.*"

6. Page 16 line 3: Is there any evidence to back this up?

Typical error diagnostics as presented in this paper allow for a quantification of (expected) systematic errors. Obviously, many assumptions have to enter into these calculations. An example for such an error diagnostic is given in Fig. 6. Comparing the magnitude of the combined systematic errors (that is all error terms except the noise one) with the retrieved volume mixing ratios shows that for the primary targets, the systematic error is mostly dominated by spectral line uncertainty, whereas the secondary targets are also strongly affected by uncertainty in background gases or have a total error (much) larger than the retrieved volume mixing ration.

If such calculations count as hard evidence lies beyond the scope of this paper.

However, during the RECONCILE campaign, comparison against other instruments for the primary retrieval targets was feasible, which asserted the validity of the retrieved volume mixing ratios and the associated error bars (actually, the errors seem to have been overestimated generally). The secondary targets could not be validated in this way, which is certainly part of the reason why they are seen as only secondary.

7. Page 16 line 19: Based on what data sets is 200 the typical scale difference between horizontal and vertical structures in the atmosphere?

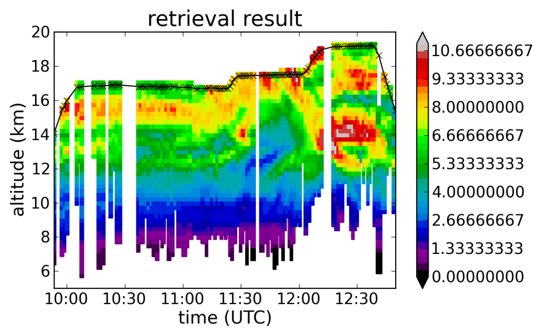
This value was derived from informal communication and should not be taken too seriously as it serves only as a starting point. We restate as "*A natural starting point for the horizontal regularisation would be an approximate scale difference between vertical and horizontal length for large meso- or synoptic-scale structures in the atmosphere, i.e. ≈ 200.*". However, according to the current limited experience with the method, perceived best values tend to lie between 100 and 400.

8. Page 16 line 24 (also, page 20 line 11): "not very pleasing to the eye..." This is not a very scientific statement. Could this be further quantified/classified?

Sect. 3.5 gives a quantitative discussion. While several "optimal" criteria exist to derive the "right" regularisation strength, these often just try to replicate the "trained eye" of experts. As such, the author holds the visual impression of line and cross-sections plots in great esteem. The human eye is a great instrument to part order and chaos. However, to help the doubting reader, a reference to the quantitative noise level discussion was added: "*... but it is visually already much more pleasing than the baseline setup (see Sect. 3.5 for a quantitative discussion).*"

9. Page 17 line 6: The author mention that they have left out a plot of the stronger HNO3 distribution, which they claim confirms the structures they managed to extract from the weaker gases thanks to the improved algorithm. This plot would be a strong evidence of their conclusions.

We do not believe that an HNO_3 plot needs to be included into the paper, as it has been published in Ungermann et al. (2012). As the published plot uses contours, we repeat it here in the same style used for the plots in the paper under discussion:



One can see the inclined structure between ≈11:40 and ≈12:10 UTC between 11 and 13 km. Due to the discrete colour scale, any quantitative comparison needs to be taken with a grain of salt, though. In the baseline $ClONO_2$ plot, the corresponding structure starts at ≈11:48 UTC. One profile has already more than 0.2 ppbv, while the one next to it shows has less than 0.2 ppbv. Durther to the right the VMRs becomes continuous. The factor-200 regularisation shows a consistent picture from 11:46 UTC onward. While not noted in the paper, the $ClONO_2$ plot with reduced vertical regularisation brings this feature out even better: It starts at 11:42 UTC and corresponds best to the HNO_3 mixing ratios.

We modify the original paper to "*Several features even look better, for example, the inclined outflow of increased* $ClONO_2$ *at 11:50* UTC *at 12* km *is now consistent over all neighbouring profiles.*" to allow a better identification of the structure related to (11:45 ↦ 11:50 and addition of "inclined").

Please note further the good correspondence of the horizontal filaments at 11:00 UTC between the baseline HNO_3 and the reduced vert. reg. strength $ClONO_2$ plot.

10. The scenario with factor-20 000 regularisation strength seems a bit extreme. Its results in this case study indicate that this would not be a viable choice for a real application. Is it worthwhile including it in the case study?

This particular regularisation strength is extreme by choice as it serves as a counterpoint to the baseline setup with no horizontal regularisation and thereby as an example of what might be too much. However, a main point here is that the retrieved VMRs are far from horizontally homogeneous (especially for CFC-11) despite the chosen strength, so Fig. 5 deserves its place.

11. Page 26 line 15: If this technique can indeed improve the retrieval of instrument parameters this would be a major strengths in its books, but this is only given as a side note here. It seems worthwhile to expand on this claim.

For the CRISTA-NF limb-sounder, only the single instrument parameter "offset" is being retrieved. When retrieving individual profiles, the retrieved offset varies quite a bit between the profiles, which is compensated for by the retrieved extinction.

Regularising the extinction gives already a rather smooth and stable offset parameter. In this case the 1-D retrieval seems to have difficulties in determining both extinction and offset from the measurements. In practise however, the retrieved trace gas volume mixing ratios are very similar in either case so that the “better” offset and extinction do not really benefit the primary targets.

However, we plan to investigate the capabilities of this technique for the newer GLORIA instrument with its more complicated 2-D detector.

12. It’s somewhat unclear how much this analysis is specifically suited to the instrument under test (i.e. CRISTA-NF), and how much it would benefit other techniques. The original premise of the study is that due to the high sampling rate of CRISTA the individual scans are horizontally correlated, a fact which is exploited with this retrieval technique. However, other limb-sounding instruments have lower sampling rates. I.e. just looking at instruments on the same air-borne platform, the other infra-red limb-sounder MIPAS-Str takes a slightly longer time to complete a full atmospheric scan, and the microwave instrument MARSCHALS even takes a significantly longer time to do so. We guess at one point the benefit of this approach becomes marginal, but it’s not quite clear what this threshold is. On a similar note, the application of this techniques to future air-borne or satellite missions is not completely clear. The GLORIA-AB infra-red limb-imager is using a truly tomographic scanning mode, so ’neighboring’ profiles are directly correlated, not just indirectly. The satellite missions will mostly be forward or rearward looking (i.e. PREMIER), so a directly tomographic retrieval approach might be more applicable in these cases. The author mentions that for pushbroom imagers the retrieval would be split up in swaths and each, in which case we presume that each swath would be subject to a tomographic retrieval, and that horizontal correlation could then be used to improve the 3D fields. We believe to understand that the technique described here could be used in a single step to retrieve 3D datasets, but this is only mentioned in a side note and it’s not quite clear that this indeed the case, nor what additional steps would be necessary to implement such an algorithm to a full 3D scenario, as compared to simpler example case of this study. Overall we perceive a certain ambiguity as to whether this is a paper documenting the next stage in the analysis of CRISTA-NF data, on which it clearly delivers, or if it’s meant to be a general paper on a new data processing algorithm, in which case more evidence to underline the relevance to other measurement techniques would be welcome. We also missed a statement whether the analysis of additional campaign data of CRISTA-NF is planned in the near future.

Obviously, the algorithm is currently being used for our retrievals for CRISTA-NF and GLORIA. One paper regarding CRISTA-NF data acquired during the AMMA campaign in 2006 is in preparation and should shortly appear in ACPD. This does not preclude it from being applied to other instruments, even though the author cannot focus his attention on those use cases. The intent of the paper is however to provide a general description of the algorithm applicable to all kind of instruments. The paper includes one specific use case to demonstrate that it is working and to analyse its benefits and drawbacks.

Existing instruments such as MARSHALS measures probably indeed too slowly to benefit from the technique, whereas MIPAS-STR might have a sufficient amount of measurements. However, newly built instruments in either frequency range will almost certainly acquire profiles faster and will therefore be able to benefit from this technique.

With respect to nadir satellite instruments, current sounders such as AIRS or IASI should be able to benefit from this technique, as the amount of taken profiles is comparable. The discussed principle is directly applicable with either a 2-D or even 3-D retrieval. A dedicated case study for nadir sounders would justify its own paper. Due to the more ill-posed nadir problem, one cannot simply transfer the results of the presented case study.

With respect to PREMIER, the reviewer grasped the concept correctly.

Obviously, there should not be doubt with respect to these topics in the conclusion. The discussion on applicability to other instruments was therefore expanded to:

Using cross-section retrievals, it is possible to produce a better representation of the true atmospheric state by exploiting the high measurement density of modern instruments and the self-similarity of the atmosphere. The better reproduction of thin vertical layers is important for the analysis of mixing processes in the upper troposphere/lower stratosphere. Especially for trace gases with weak signature, the technique reduces the image noise significantly without noticeable degradation of the horizontal resolution. The algorithm is therefore currently used to process further CRISTA-NF data (Ungermann et al., 2012b) and initial (non-tomographic) measurements by GLORIA (Gimballed Limb Observer for Radiance Imaging of the Atmosphere; see Ungermann et al., 2010). This technique might also be used to more reliably derive constant and slowly varying instrument parameters, which cannot be determined from pre- or post flight calibration. But the technique should also be applicable to older airborne sounders with a more sparse sampling such as MIPAS-STR (e.g. Woiwode et al., 2012), albeit with less resulting image enhancements.

In contrast to the title of this paper, which was chosen mostly due to the discussed use-case, it is straightforward to extend the presented technique to retrievals for current satellite-borne nadir-sounders, as these instruments also measure closely spaced profiles (e.g. both the Atmospheric InfraRed Sounder (Aumann et al., 2003) and the Infrared Atmospheric Sounding Interferometer (Clerbaux et al., 2009) have a (sub)pixel size of 12 to 13.5 km*; the operational retrievals for both instruments combine sub-pixels to reduce measurement noise, which could be achieved with less cost* *of horizontal resolution with the proposed algorithm). As the viewing geometry is different, the technique needs to be slightly adapted. Instead of cross-sections, 3-D cubes would be retrieved with added horizontal regularisation in both along-flightpath and across-flightpath direction. This only changes the state vector representation and the setup of the regularisation matrix. While the resulting problem size would be noticeably larger than in the presented cross-section retrieval, it is not larger than tomographic problems already treated by Ungermann et al. (2011). The technique might be even more beneficial when applied to nadir-sounders due to the greater ill-posedness of the retrieval compared to limb-sounder retrievals. Examining and quantifying this in detail deserves further study.*

It is straightforward to extend the presented technique also to proposed near-future satellite limb-imagers (Riese et al., 2005; ESA, 2012). Such a rearward-looking instrument uses a 2-D detector to acquire multiple profiles simultaneously. Due to the high measurement speed, consecutive images overlap in the sense that they measure largely the same airmass. Assembling the measured profiles into several 2-D swaths parallel to the flight-path allows the use of 2-D tomographic retrieval techniques (Carlotti et al., 2001; Ungermann et al., 2010a, e.g.) to achieve an excellent resolution in all three dimensions. The proposed technique is capable of evaluating all 2-D swaths together in a single 3-D tomographic retrieval. By exploiting the similarity between neighbouring swaths, the described technique would stabilise the inherently more ill-posed tomographic retrieval problem and reduce the image noise level while possibly also improving the resolution. This might enhance the scientific capabilities of the limb-imager, for example with respect to gravity wave detection (see Preusse et al., 2009).

2.2 Technical Corrections

1. Page 24 lines 4-6: Whole sentence is unclear, please rephrase.

The section was rephrased as *However, a closer analysed reveals that the matrices being multiplied and inverted in Eq. (15) are not too well conditioned to begin with (about* $\approx 10^5$ *for the summed matrices after symmetrically scaling the diagonal to 1) and both matrix-matrix multiplication and matrix-inversion are rather sensitive to high condition numbers: for the given matrices, the combination of the squaring and inversion in Eq. (15) introduces sufficient error to effectively remove* $\approx$*33 bit of information, which is more than is usually employed for storing the covariance matrices.*

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