Author comment regarding the assimilation method

We thank the referee for the comments. The referee raises concerns regarding the validity of our inversion method. However, we are convinced that this is indeed a misunderstanding, caused by differing conventions within the fields of data assimilation and inverse modelling. In the following, we aim to address the question regarding validity of the assimilation method; we will address the specific comments (1-4) in the final response provided along with submission of the revised manuscript.

In our inversion, contrary to numerical weather prediction, the observations are assimilated in a single window covering all 21 days. We realise that this aspect may not have been spelled out clearly enough in the manuscript, as it is only mentioned implicitly in section 3.1 (line 133: "state vector x is taken collectively for all time steps"). This important point will be stated upfront in the revised manuscript.

Little SO_2 was observed at the beginning of the assimilation window, which coincided with inactive phase of the eruption. It is therefore reasonable to assume that the SO_2 concentrations within the assimilation window depended only on the emission flux $\mathbf{f}$, given as a function of time and altitude. In this case, the cost function does not include a term for the background state $\mathbf{x_b}$, and instead, the possible a priori constraints are given for the emission $\mathbf{f}$:

$$\mathcal{J}(\mathbf{f}) = \frac{1}{2}(\mathbf{y} - \mathcal{H}(\mathbf{x}))^T \mathbf{R}^{-1} (\mathbf{y} - \mathcal{H}(\mathbf{x})) + \mathcal{T}(\mathbf{f}) \quad (1)$$

where $\mathbf{x} = \mathcal{M}(\mathbf{f})$ includes the model state on all time steps of the assimilation window, $\mathcal{H}$, $\mathbf{y}$, and $\mathbf{R}$ denote the observation operator, observation data, and observation error covariance matrix, and $\mathcal{T}(\mathbf{f})$ is a penalty functional. Most of the existing studies on inverting volcanic emissions have used a cost function similar to Eq. (1) with various forms of $\mathcal{T}(\mathbf{f})$, including an a priori emission source with a prescribed prior error covariance matrix (Eckhardt et al. (2008) and the subsequent works using the FLEXPART model), or Tikhonov regularisation with first or second order smoothness constraints (Boichu et al., 2013; Lu et al., 2016). Since there is little prior information on smoothness of volcanic emissions, we chose the arguably simplest option of zeroth order Tikhonov regularisation (Eq. 9 in the manuscript) as the point of reference.

A limitation of inversions based on Eq. (1) is that the model is taken as a strong constraint i.e., model errors developing within the assimilation window are not included in the cost function. Since the model errors are not negligible, their effect on the residual $\mathbf{y} - \mathcal{H}(\mathbf{x})$ needs to be included into the observation error covariance matrix $\mathbf{R}$. Also this aspect has been recognized by previous flux inversion studies for both volcanic (Seibert et al., 2011; Stohl et al., 2011) and other emissions (eg. Bergamaschi et al. (2005); Bocquet, (2012)).

It would be possible to arrange the inversion into successive assimilation windows as done by Elbern et al. (2007) in context of air quality forecasting. In this case, the initial state should probably be included in the control vector; however, this would not remove the assumption of negligible model errors within the assimilation window. This approach is so far untested for volcanic emissions – Flemming and Inness (2013) estimated the emission fluxes in a separate step.

The reviewer suggests that the truncated iteration is only a way to "prevent the system from wandering too far", with which we respectfully disagree. Truncated iteration (often simply called iterative regularisation) has been shown to be a computationally efficient method for regularising large-scale inverse problems, and it is discussed extensively in textbooks (e.g. Hansen, 2010; Kaipio and Somersalo, 2006) in addition to the papers (Calvetti et al., 2002; Fleming, 1990; Kilmer and O'Leary, 2001) cited in the manuscript.

The behaviour of RMSE as a function of iteration number is therefore not an error, but fully consistent with what is expected for many iterative methods when applied to an ill-posed problem. As seen in the left panels of Figs. 3 and 4, the residual indeed decreases as the iteration proceeds. The eventual growth of RMSE, however, is an indication of overfitting the solution to noise. The noise will eventually contaminate the converged solution, whereas the early iterates are controlled by more robust low-frequency features of the data. Truncating the iteration prevents the overfitting and results in a more accurate solution than that obtained by allowing the iteration to converge.

Although the self-regularising properties of iterative methods are rarely exploited in data assimilation, in our experiments the truncated L-BFGS-B iteration yielded solutions that were, depending on the point of truncation, similar or better in RMSE than those given by optimally tuned Tikhonov regularisation. With Tikhonov regularisation (Eq. 9), the iteration converges, although with weak regularisation (small α^2), the convergence becomes very slow.

Due to the reasons explained above, the tests with J_B and J_R as suggested in the review are not actually possible. However, we can use the χ^2 condition to evaluate the consistency of the $\mathbf{R}$ matrix. For a well-specified inversion, linear regression theory implies (e.g. Tarantola, 2005) that the expectation of the unregularised cost function (Eq. 2 in the manuscript) at the minimum is

$$E(J(\mathbf{f}_{opt})) = \frac{n-p}{2} \qquad (2)$$

where n is number of observations and p is number of estimated parameters. For the Eyjafjalljökull inversion with assimilation of total column and plume height (centre of mass) retrievals, $p \sim 1400$, $n \sim 1.4 \cdot 10^6$ and $J(\mathbf{f}_{opt}) \sim 6 \cdot 10^4$ with $\mathbf{f}_{opt}$ given by the L-curve criterion. The apparent discrepancy is explained by the large proportion of zero observations with only small contribution to the total cost function. If the cost function is evaluated using only observations corresponding to positive SO_2 detections, we have $n_{y>0} \sim 10^5$ and $J_{y>0}(\mathbf{f}_{opt}) \sim 5 \cdot 10^4$. Even if Eq. (2) does not strictly hold for subsets of observations, this suggests that our $\mathbf{R}$ is reasonably specified, especially given that the assumed values for $\mathbf{R}_{\mathbf{model}}$ are at best a crude approximation for the actual model uncertainty.

References

Bergamaschi, P., Krol, M., Dentener, F., Vermeulen, A., Meinhardt, F., Graul, R., Ramonet, M., Peters, W., Dlugokencky, E.J., 2005. Inverse modelling of national and European CH4 emissions using the atmospheric zoom model TM5. Atmos. Chem. Phys. 5, 2431–2460. doi:10.5194/acpd-5-1007-2005

Bocquet, M., 2012. Parameter-field estimation for atmospheric dispersion: application to the Chernobyl accident using 4D-Var. Q. J. R. Meteorol. Soc. 138, 664–681. doi:10.1002/qj.961

Boichu, M., Menut, L., Khvorostyanov, D., Clarisse, L., Clerbaux, C., Turquety, S., Coheur, P.-F., 2013. Inverting for volcanic SO2 flux at high temporal resolution using spaceborne plume imagery and chemistry-transport modelling: the 2010 Eyjafjallajökull eruption case study. Atmos. Chem. Phys. 13, 8569–8584. doi:10.5194/acp-13-8569-2013

Calvetti, D., Lewis, B., Reichel, L., 2002. GMRES, L-curves, and discrete ill-posed problems. BIT Numer. Math. 42, 44–65.

Eckhardt, S., Prata, A.J., Seibert, P., Stebel, K., Stohl, A., 2008. Estimation of the vertical profile of sulfur dioxide injection into the atmosphere by a volcanic eruption using satellite column measurements and inverse transport modeling. Atmos. Chem. Phys. 8, 3881–3897. doi:10.5194/acpd-8-3761-2008

Elbern, H., Strunk, A., Schmidt, H., Talagrand, O., 2007. Emission rate and chemical state estimation by 4-dimensional variational inversion. Atmos. Chem. Phys. 7, 3749–3769. doi:10.5194/acpd-7-1725-2007

Fleming, H.E., 1990. Equivalence of regularization and truncated iteration in the solution of III-posed image reconstruction problems. Linear Algebra Appl. 130, 133–150. doi:10.1016/0024-3795(90)90210-4

Flemming, J., Inness, A., 2013. Volcanic sulfur dioxide plume forecasts based on UV-satellite retrievals for the 2011 Grímsvötn and the 2010 Eyjafjallajökull eruption. J. Geophys. Res. Atmos. 118. doi:10.1002/jgrd.50753

Hansen, P.C., 2010. Discrete Inverse Problems: Insight and Algorithms, Fundamentals of Algorithms. Society for Industrial and Applied Mathematics.

Kaipio, J., Somersalo, E., 2006. Statistical and Computational Inverse Problems, Applied Mathematical Sciences. Springer New York.

Kilmer, M.E., O'Leary, D.P., 2001. Choosing Regularization Parameters in Iterative Methods for Ill-Posed Problems. SIAM J. Matrix Anal. Appl. 22, 1204–1221. doi:10.1137/S0895479899345960

Lu, S., Lin, H.X., Heemink, A.W., Fu, G., Segers, A.J., 2016. Estimation of Volcanic Ash Emissions Using Trajectory-Based 4D-Var Data Assimilation. Mon. Weather Rev. 144, 575–589. doi:10.1175/MWR-D-15-0194.1

Seibert, P., Kristiansen, N.I., Richter, A., Eckhardt, S., Prata, A.J., Stohl, A., 2011. Uncertainties in the inverse modelling of sulphur dioxide eruption profiles. Geomatics, Nat. Hazards Risk 2, 201–216. doi:10.1080/19475705.2011.590533

Stohl, A., Prata, A.J., Eckhardt, S., Clarisse, L., Durant, A., Henne, S., Kristiansen, N.I., Minikin, A., Schumann, U., Seibert, P., Stebel, K., Thomas, H.E., Thorsteinsson, T., Tørseth, K., Weinzierl, B., 2011. Determination of time- and height-resolved volcanic ash emissions and their use for quantitative ash dispersion modeling: the 2010 Eyjafjallajökull eruption. Atmos. Chem. Phys. 11, 4333–4351. doi:10.5194/acp-11-4333-2011

Tarantola, A., 2005. Inverse problem theory and methods for model parameter estimation. Society of Industrial and Applied Mathematics.