Using an offline ensemble Kalman filter with in situ and satellite measurements to estimate ammonia emissions in the Netherlands

Simeng Li[1], Enrico Dammers[1,2], Beatriz Herrera[2], Arjo Segers[2], Mark W. Shephard[3], Martin Van Damme[4,5], Lieven Clarisse[4], Shelley van der Graaf[6], Roy Wichink Kruit[6], and Jan Willem Erisman[1]

[1]Institute of Environmental Sciences, Universiteit Leiden, Leiden, the Netherlands
[2]Air Quality and Emissions Research, Netherlands Organisation for Applied Scientific Research (TNO), Utrecht, The Netherlands
[3]Environment and Climate Change Canada, Toronto, Ontario, Canada
[4]Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing (SQUARES), BLU–ULB Space Research Center, Université Libre de Bruxelles (ULB), Brussels, Belgium
[5]Royal Belgian Institute for Space Aeronomy, Brussels, Belgium
[6]Rijksinstituut voor Volksgezondheid en Milieu, RIVM, Bilthoven, the Netherlands

Correspondence: Simeng Li (s.li@cml.leidenuniv.nl)

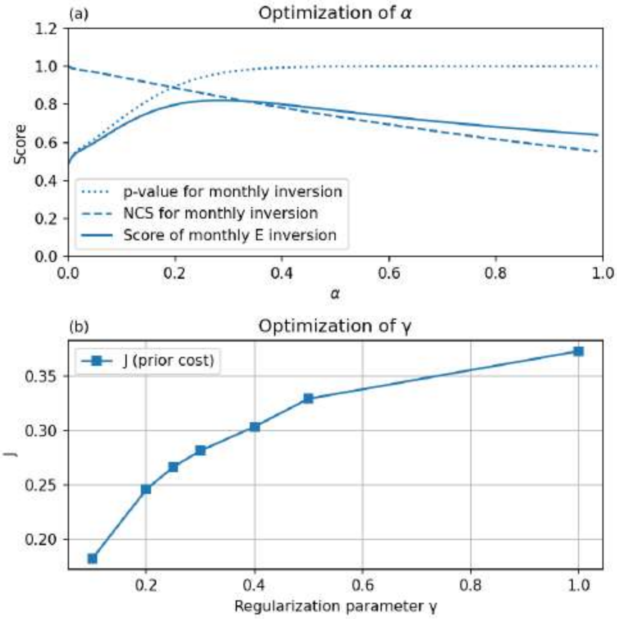


Figure S1. The optimisation of α and γ for the MAN observation. $\alpha = 0.3$ and $\gamma = 1.0$ is applied in this study.

Table S1. Optimised regularisation parameters (γ) for satellite data assimilation by month and observation configuration. Values were determined through $\mathcal{J}_A \approx n$. Smaller values indicate stronger regularisation, applied in months with larger observation counts or higher retrieval uncertainty. For in situ observations, $\gamma = 1.0$ is used throughout.

Month	CrIS-only	IASI-only	CrIS+IASI
January	0.4	0.3	0.2
February	0.1	0.1	0.4
March	0.01	0.1	0.4
April	0.01	0.05	0.1
May	0.01	0.1	0.01
June	0.01	0.1	0.02
July	0.01	0.05	0.05
August	0.01	0.05	0.02
September	0.01	0.05	0.1
October	0.1	0.1	0.4
November	0.5	0.3	0.3
December	0.3	0.3	0.2

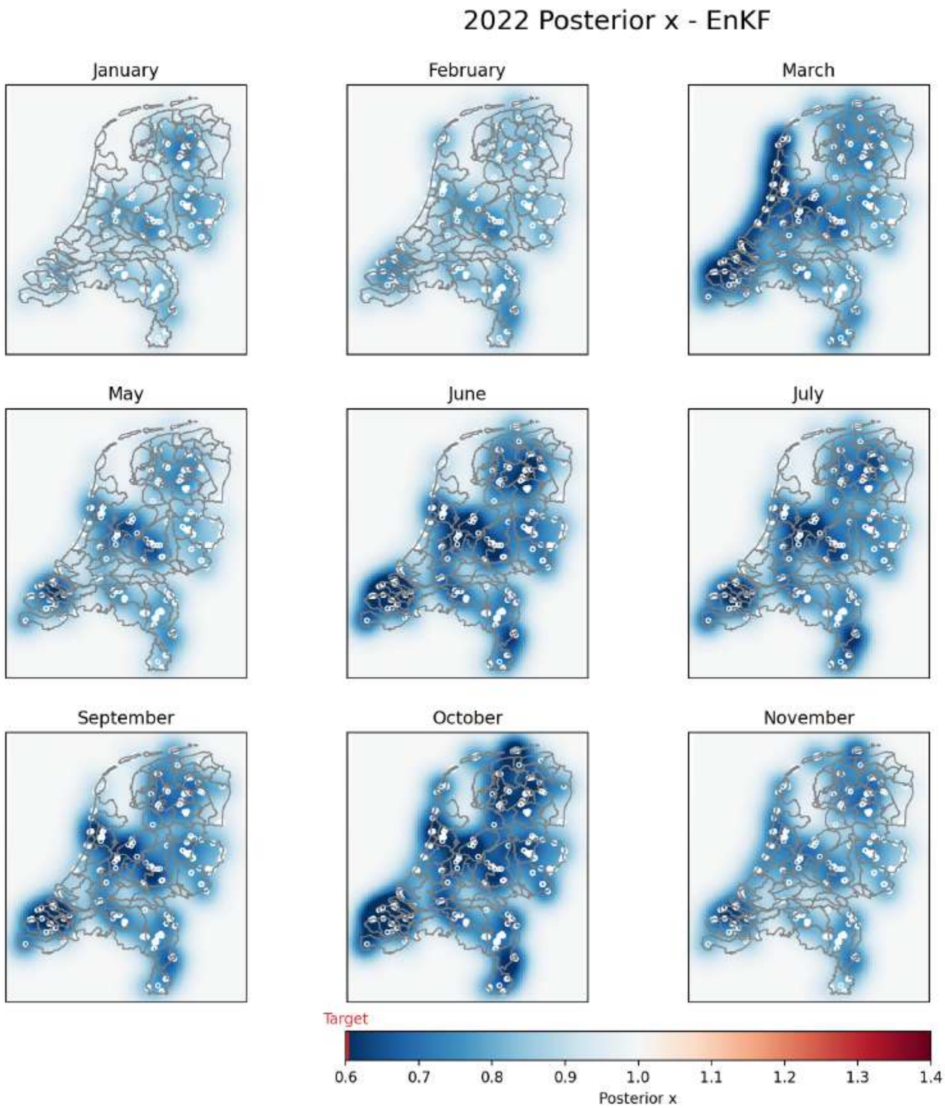


Figure S2. Monthly posterior emission scaling factors for 2022 from the uniform 40% emission reduction experiment using MAN observations only. The white dots indicate MAN station locations. Blue colors indicate emission reductions (scaling factors <1.0), with the target value of 0.6 shown by the colorbar.

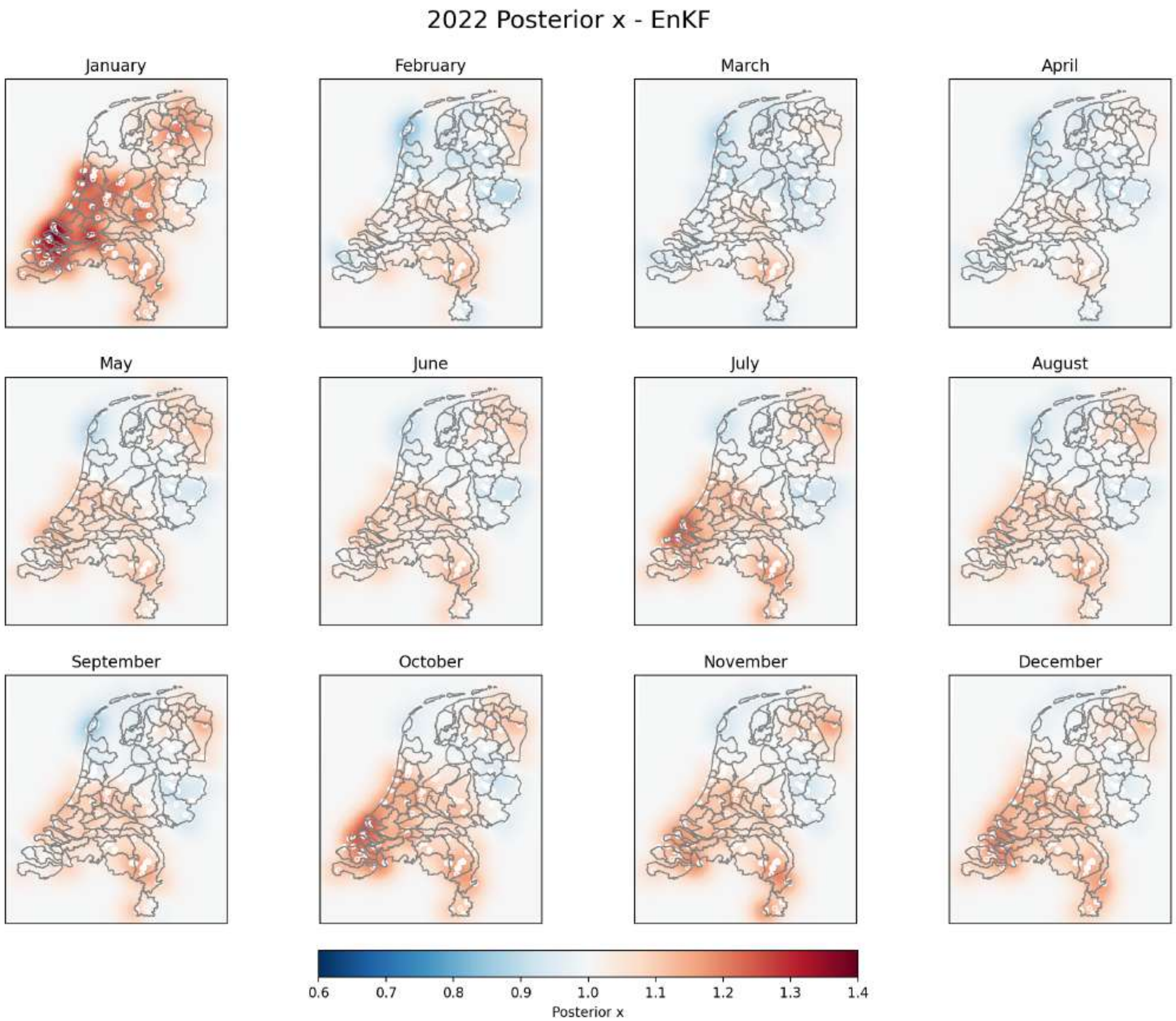


Figure S3. Monthly posterior emission scaling factors for 2022 from the multi-perturbation experiment using MAN observations only. The target spatial pattern is shown in the article, with regional perturbations ranging from 0.6 (emission reductions) to 1.4 (emission increases).

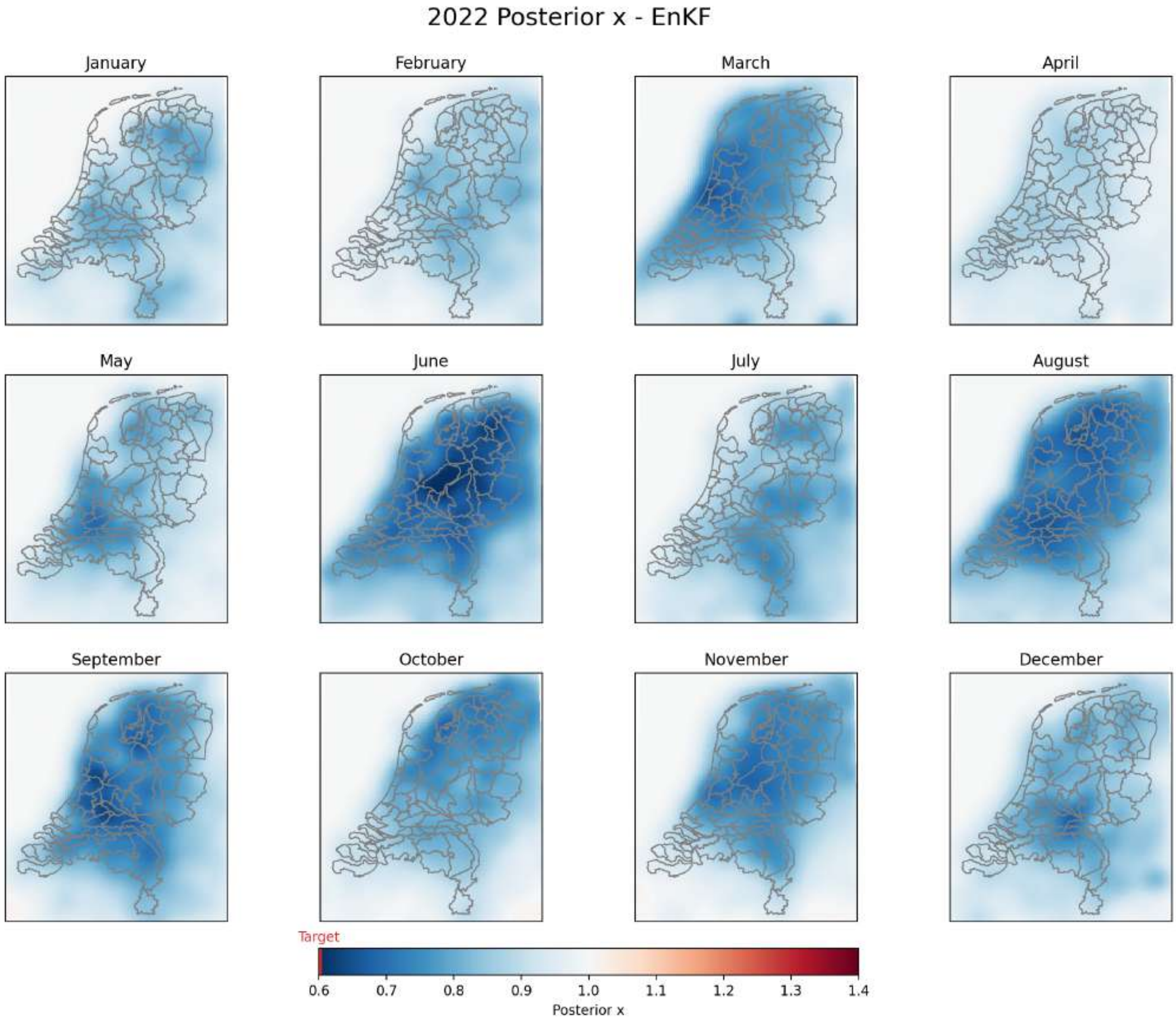


Figure S4. Monthly posterior emission scaling factors for 2022 from the uniform 40% emission reduction experiment using synthetic CrIS satellite observations only. Blue colors indicate emission reductions (scaling factors <1.0), with the target value of 0.6 shown by the colorbar.

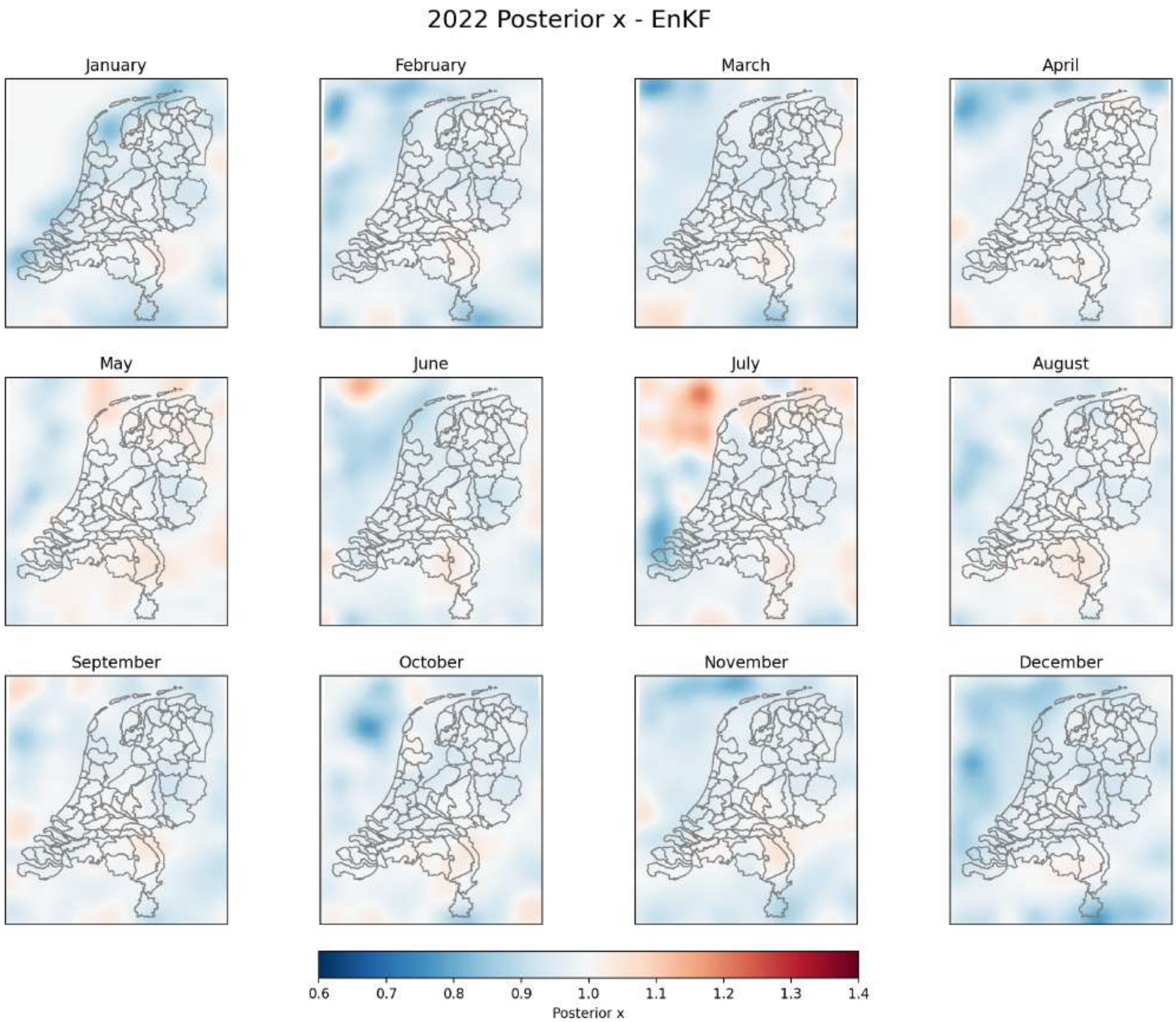


Figure S5. Monthly posterior emission scaling factors for 2022 from the multi-perturbation experiment using synthetic CrIS satellite observations only. The target spatial pattern is shown in the article, with regional perturbations ranging from 0.6 (emission reductions) to 1.4 (emission increases).

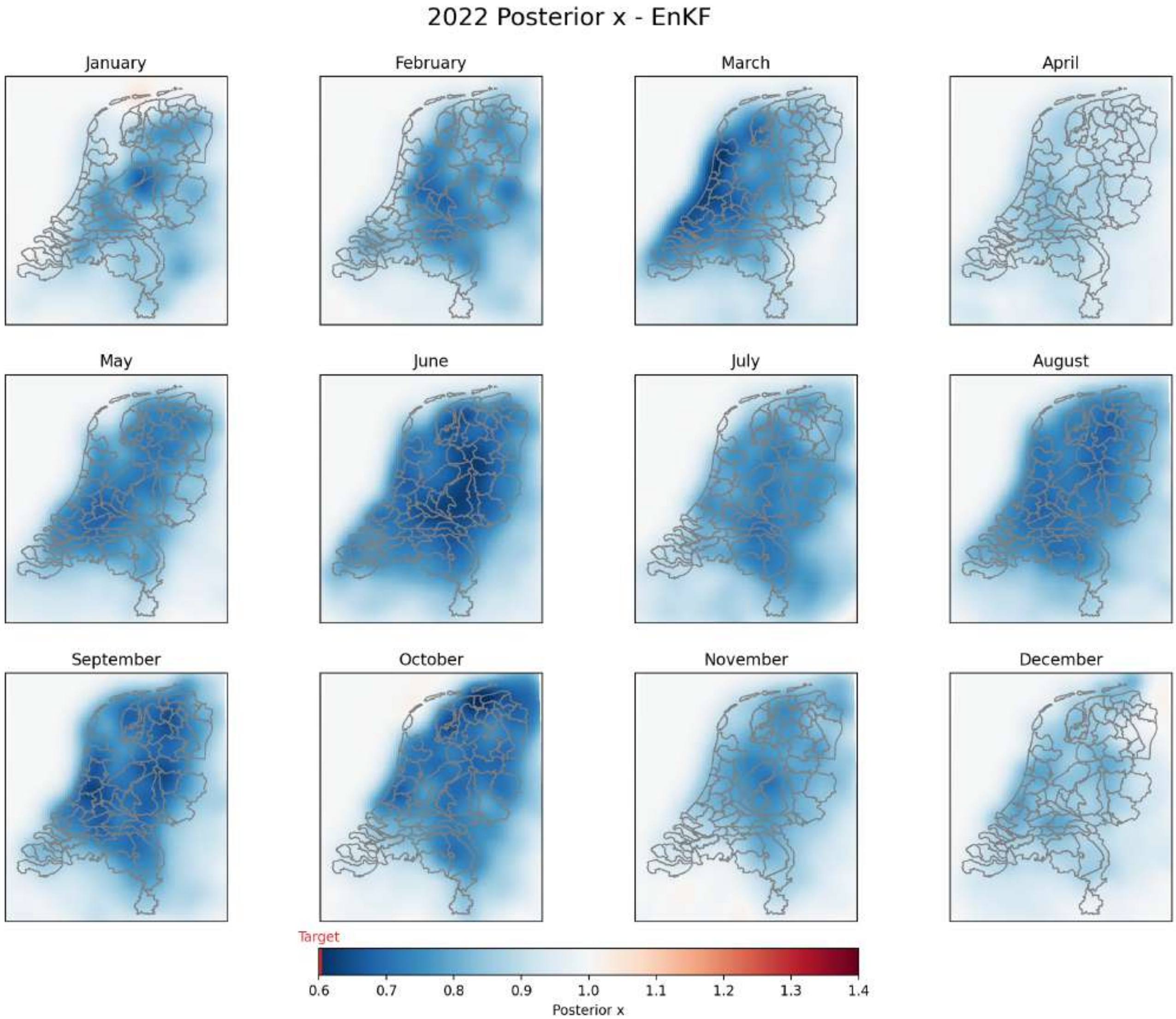


Figure S6. Monthly posterior emission scaling factors for 2022 from the uniform 40% emission reduction experiment using synthetic IASI satellite observations only. Blue colors indicate emission reductions (scaling factors <1.0), with the target value of 0.6 shown by the colorbar.

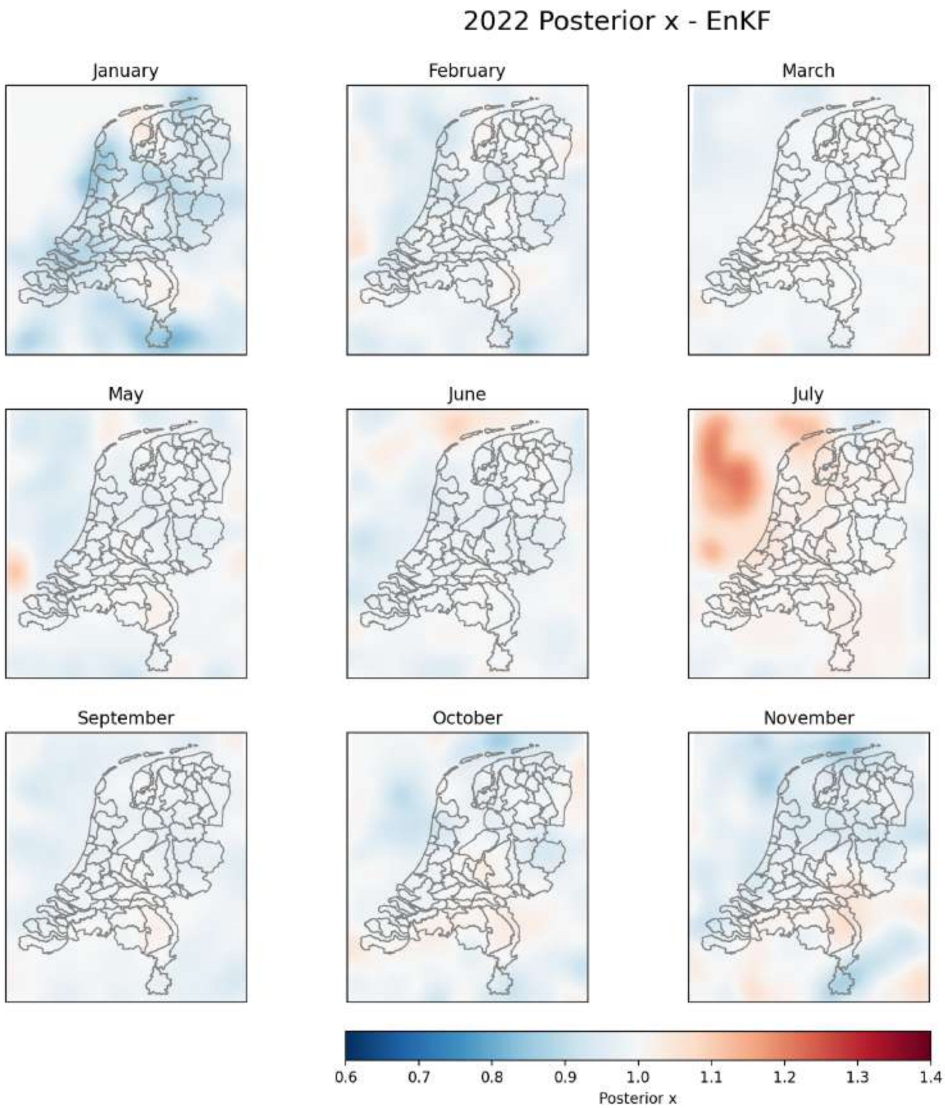


Figure S7. Monthly posterior emission scaling factors for 2022 from the multi-perturbation experiment using synthetic IASI satellite observations only. The target spatial pattern is shown in the article, with regional perturbations ranging from 0.6 (emission reductions) to 1.4 (emission increases).

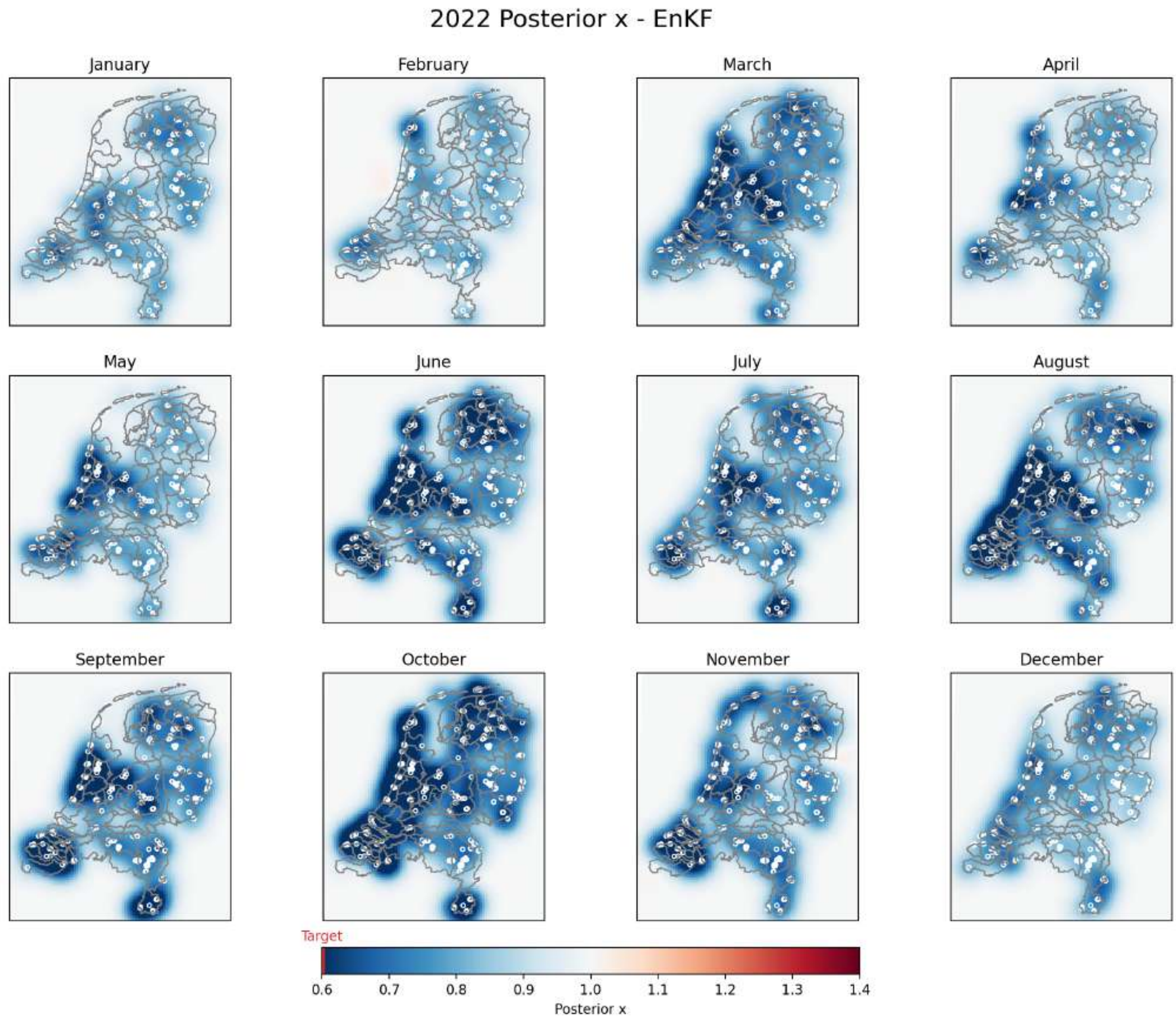


Figure S8. Monthly posterior emission scaling factors for 2022 from the uniform 40% emission reduction experiment using in situ observations (MAN+LML). The white dots indicate in situ station locations. Blue colors indicate emission reductions (scaling factors <1.0), with the target value of 0.6 shown by the colorbar.

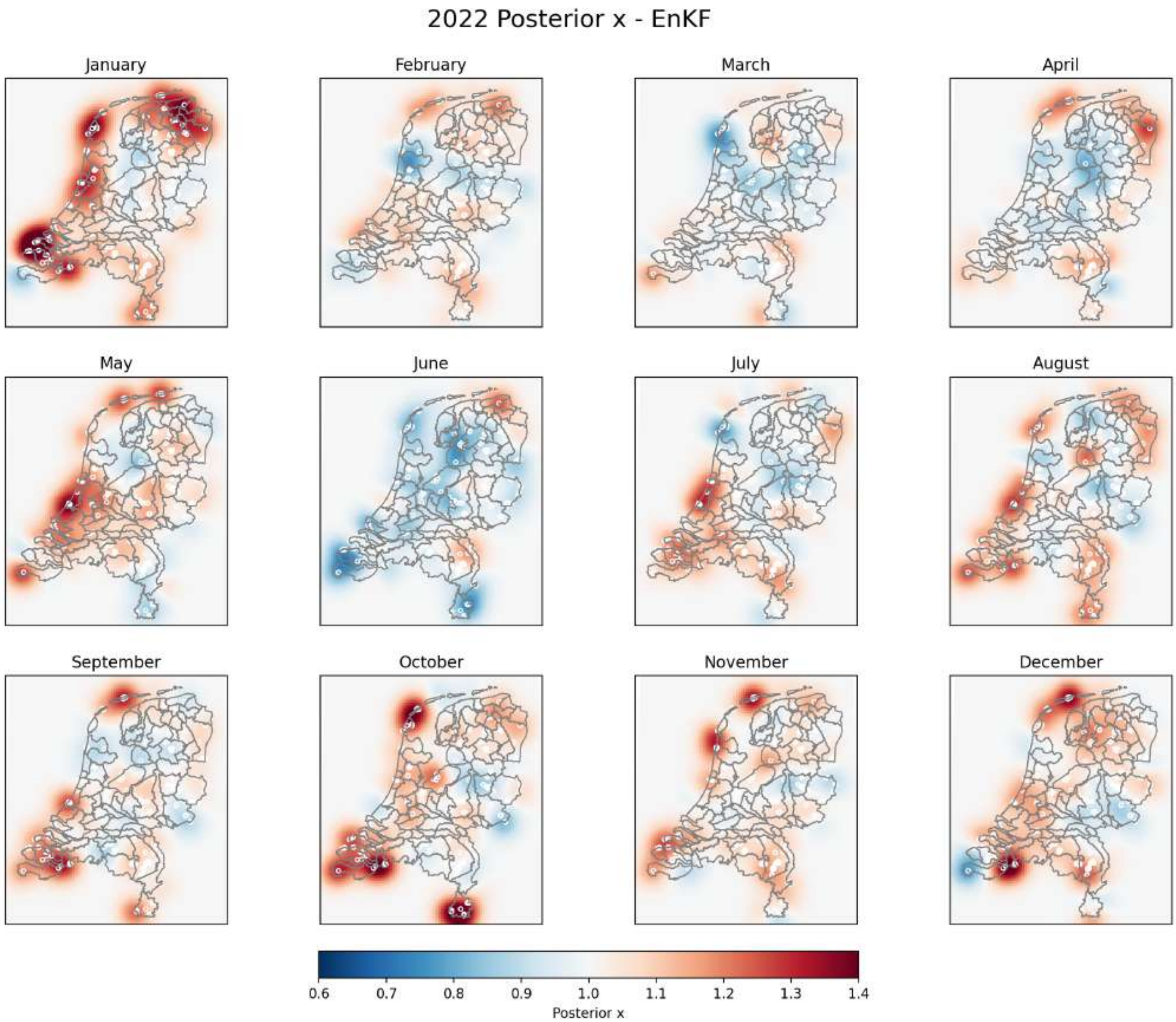


Figure S9. Monthly posterior emission scaling factors for 2022 from the multi-perturbation experiment using combined in situ measurements (MAN+LML). The white dots indicate in situ station locations. The target spatial pattern is shown in Fig. **??**, with regional perturbations ranging from 0.6 (emission reductions) to 1.4 (emission increases). **[it would be helpful to have Fig 2 close to this Figure. That makes comparison moer easy.]**

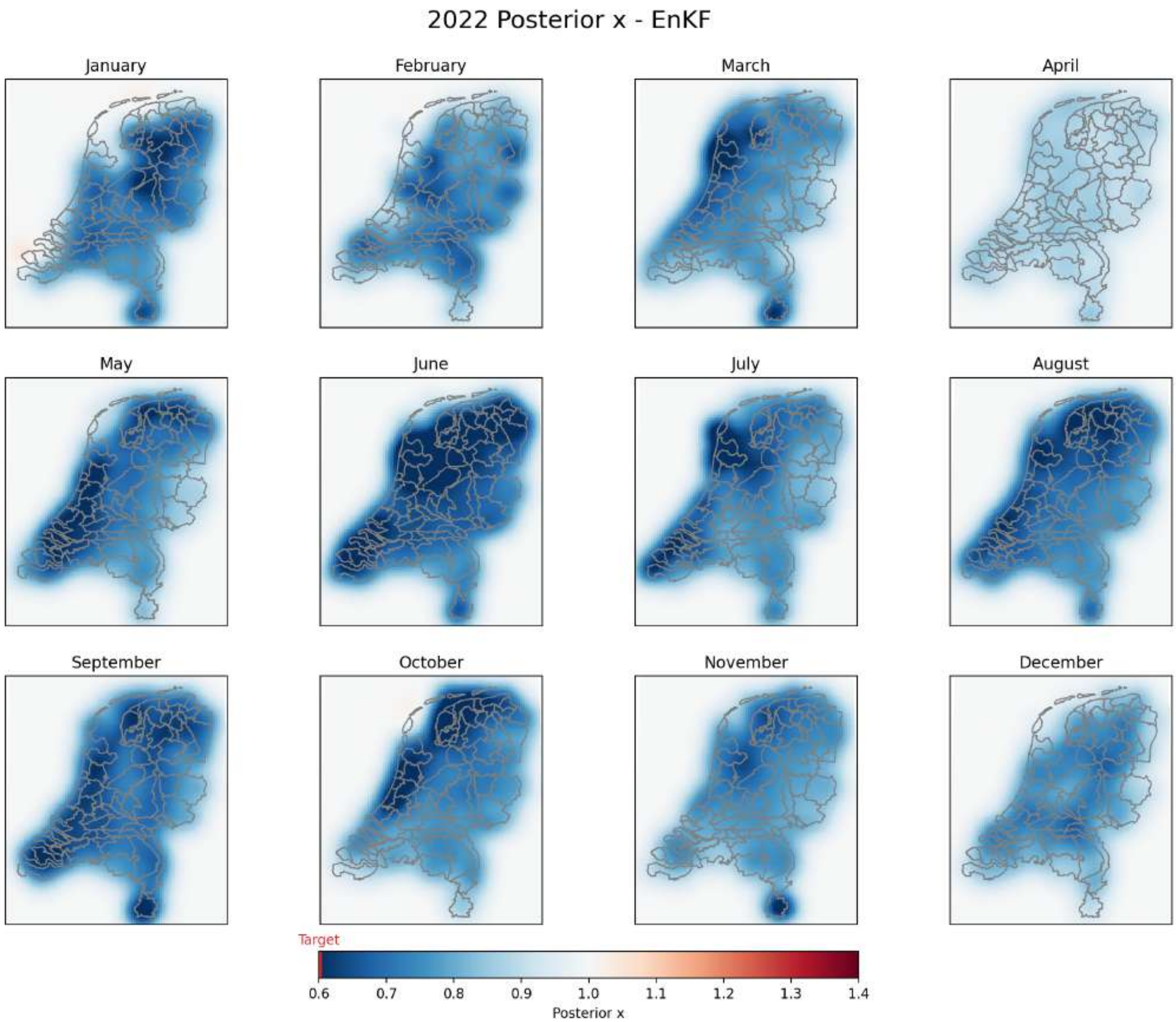


Figure S10. Monthly posterior emission scaling factors for 2022 from the uniform 40% emission reduction experiment using combined CrIS and IASI satellite observations. Blue colors indicate emission reductions (scaling factors <1.0), with the target value of 0.6 shown by the colorbar.

Figure S11. Monthly posterior emission scaling factors for 2022 from the multi-perturbation experiment using combined CrIS and IASI satellite observations. The target spatial pattern is shown in Fig. **??**, with regional perturbations ranging from 0.6 (emission reductions) to 1.4 (emission increases).

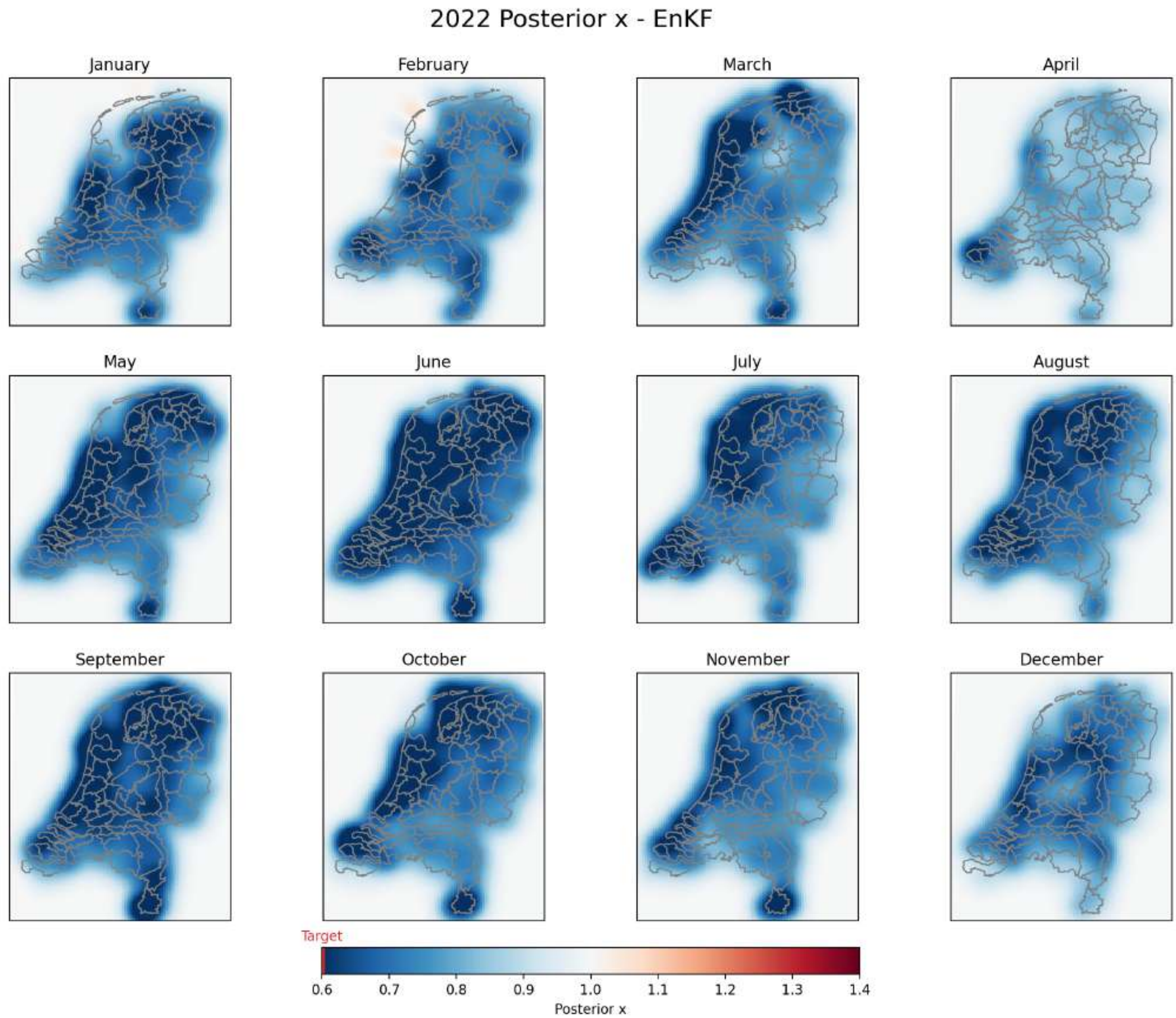


Figure S12. Monthly posterior emission scaling factors for 2022 from the uniform 40% emission reduction experiment using combined in situ (MAN+LML) and satellite (CrIS +IASI) observations. Blue colors indicate emission reductions (scaling factors <1.0), with the target value of 0.6 shown by the colorbar.

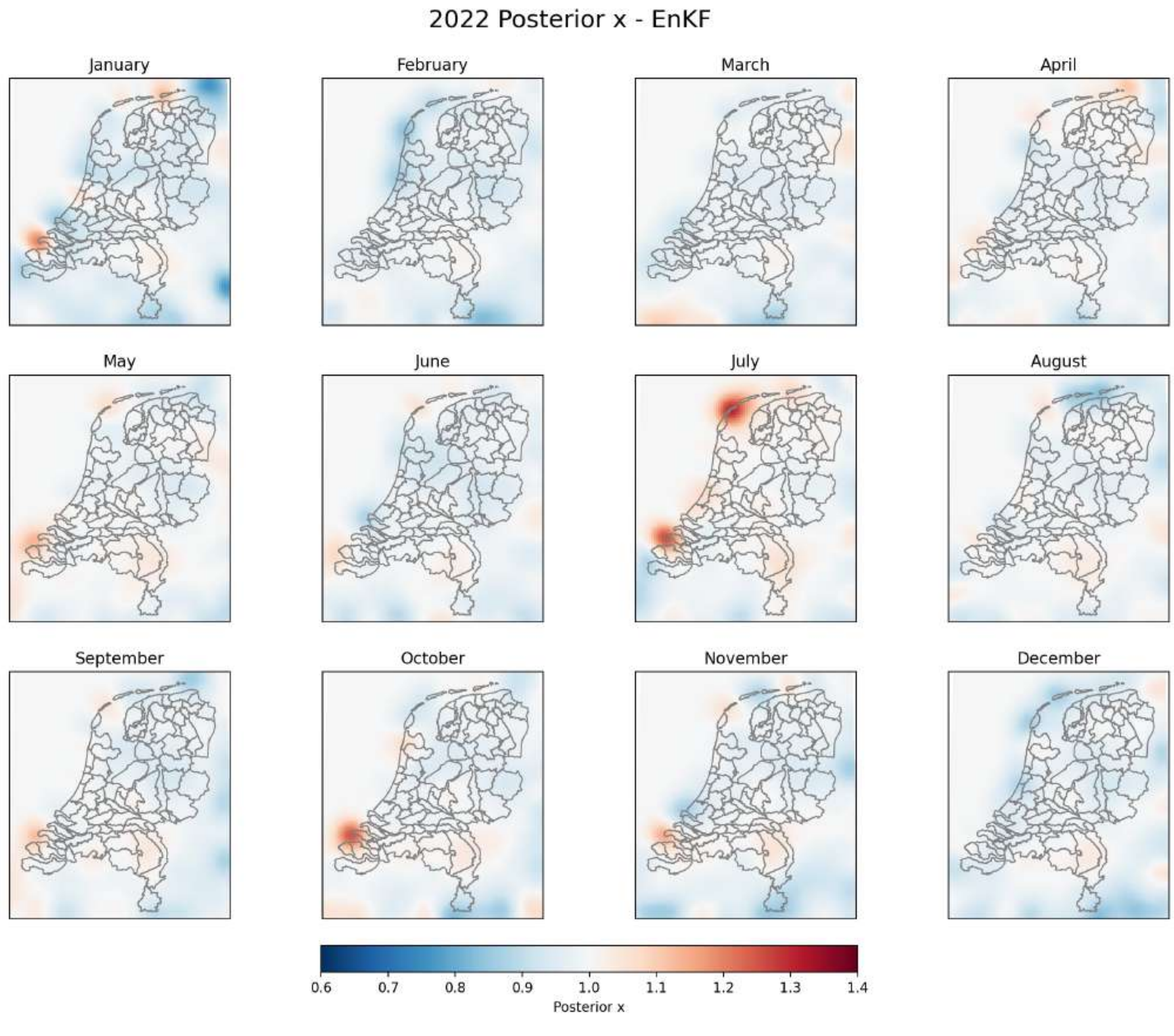


Figure S13. Monthly posterior emission scaling factors for 2022 from the multi-perturbation experiment using combined insitu (MAN+LML) and satellite (CrIS +IASI) observations. The target spatial pattern is shown in Fig. **??**, with regional perturbations ranging from 0.6 (emission reductions) to 1.4 (emission increases).

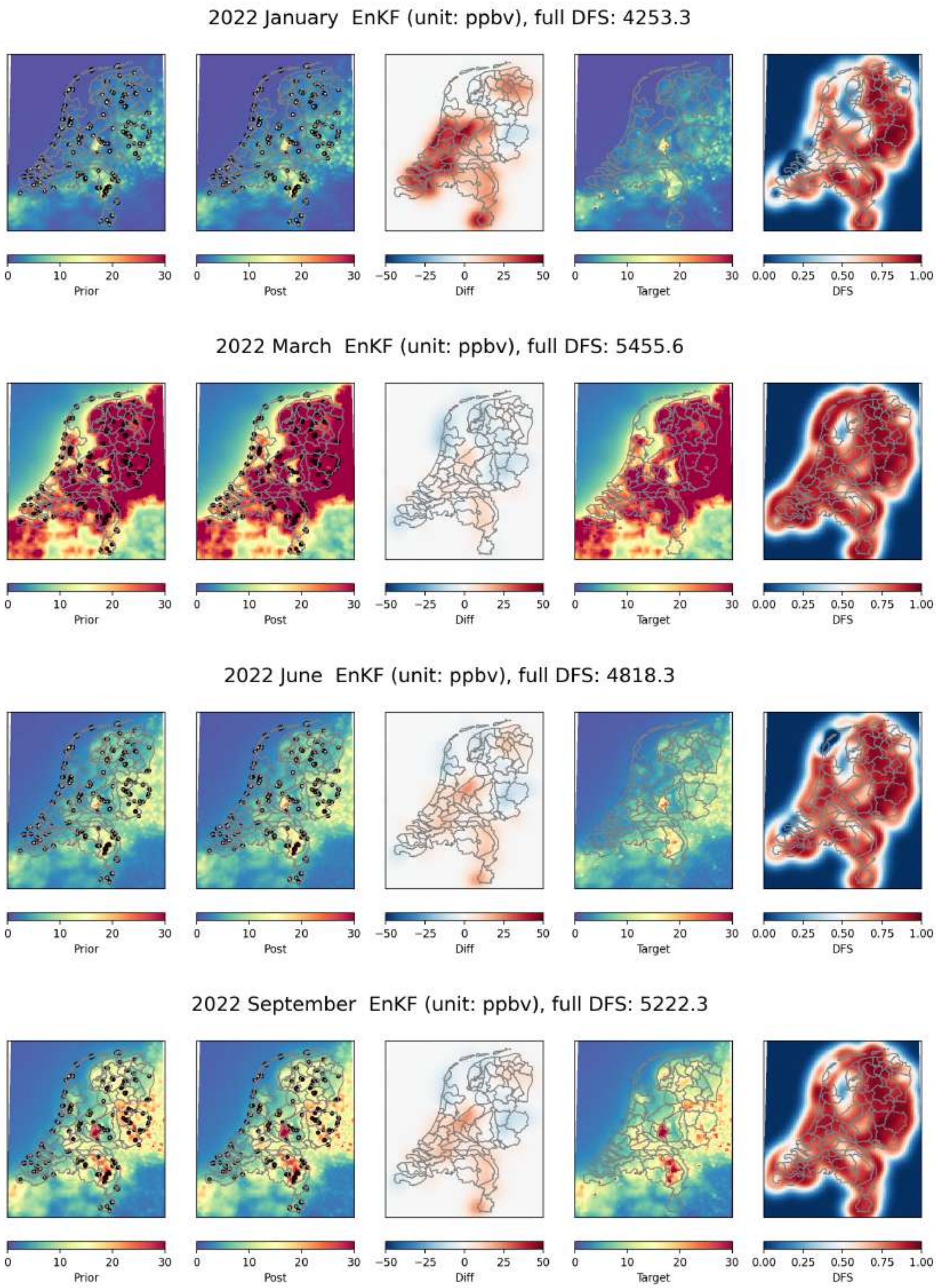
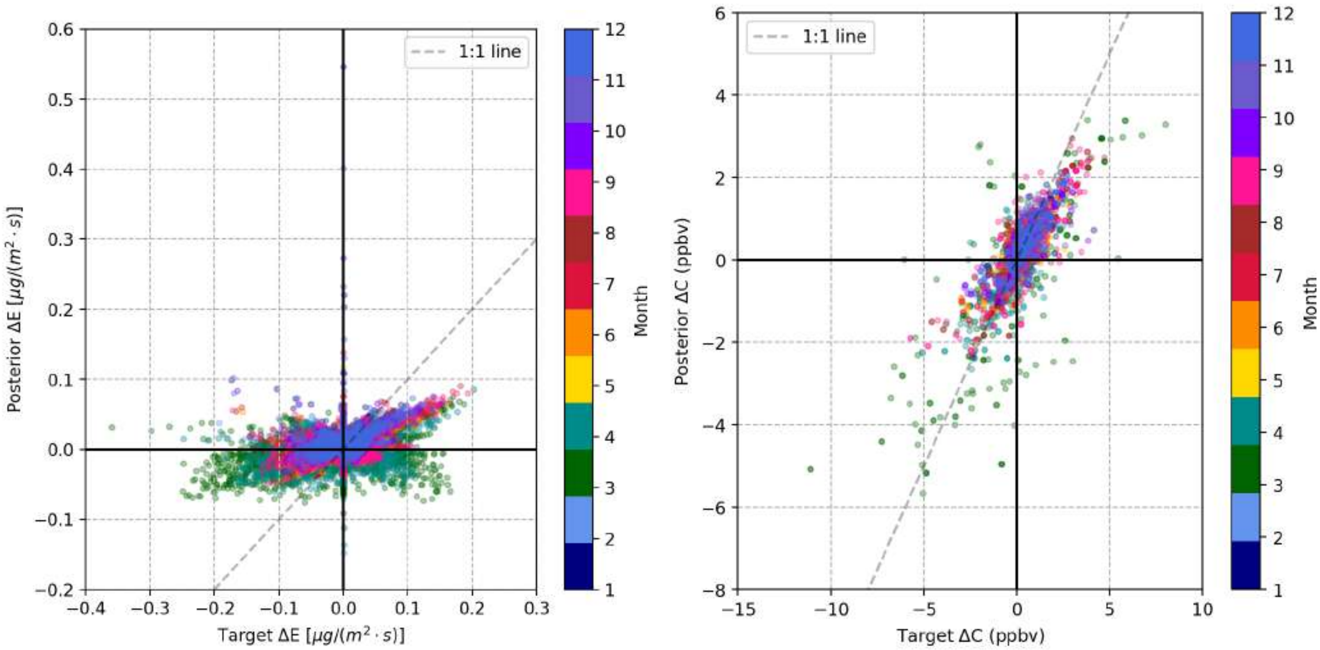


Figure S14. Monthly posterior surface concentrations for 2022 from the multi-perturbation experiment using MAN observations only. The target spatial pattern of the prior emission is shown in the article.



(a) Emission

(b) Concentration at MAN sites

Figure S15. The scatterplot comparison of monthly posterior for 2022 from the multi-perturbation experiment using MAN observations only.

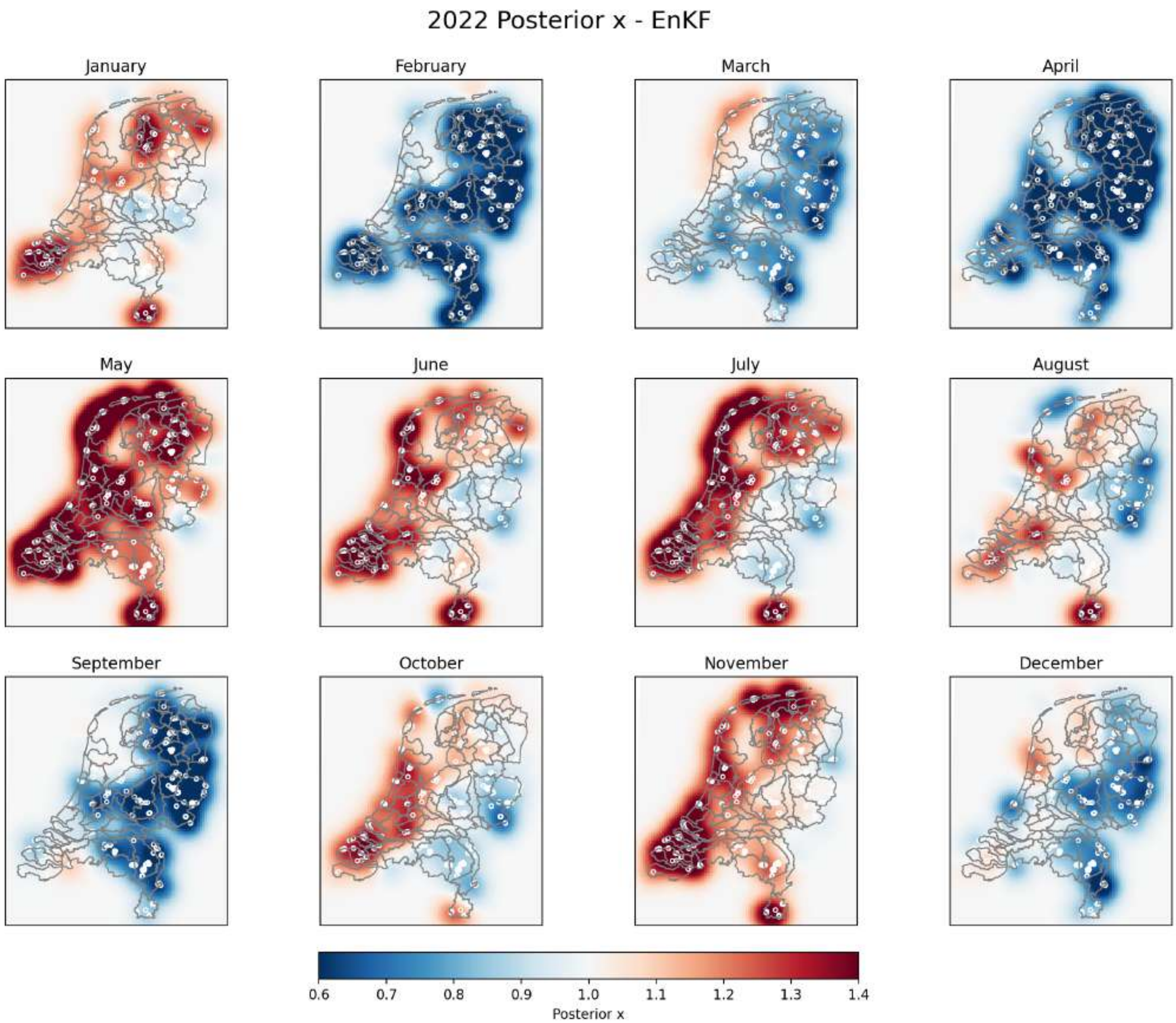


Figure S16. Monthly posterior emission scaling factors for 2022 using real MAN observations only.

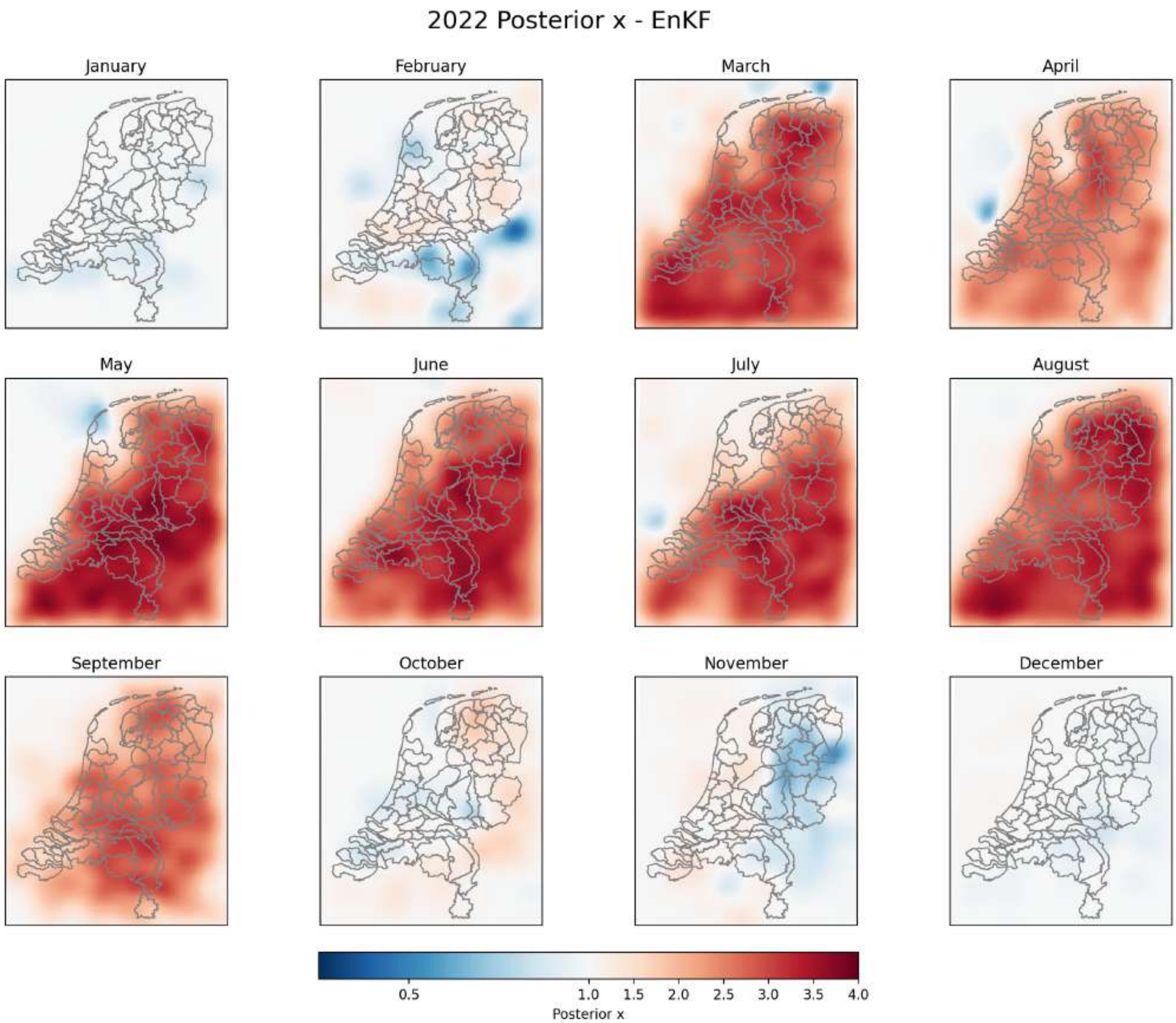


Figure S17. Monthly posterior emission scaling factors for 2022 using real CrIS satellite only.

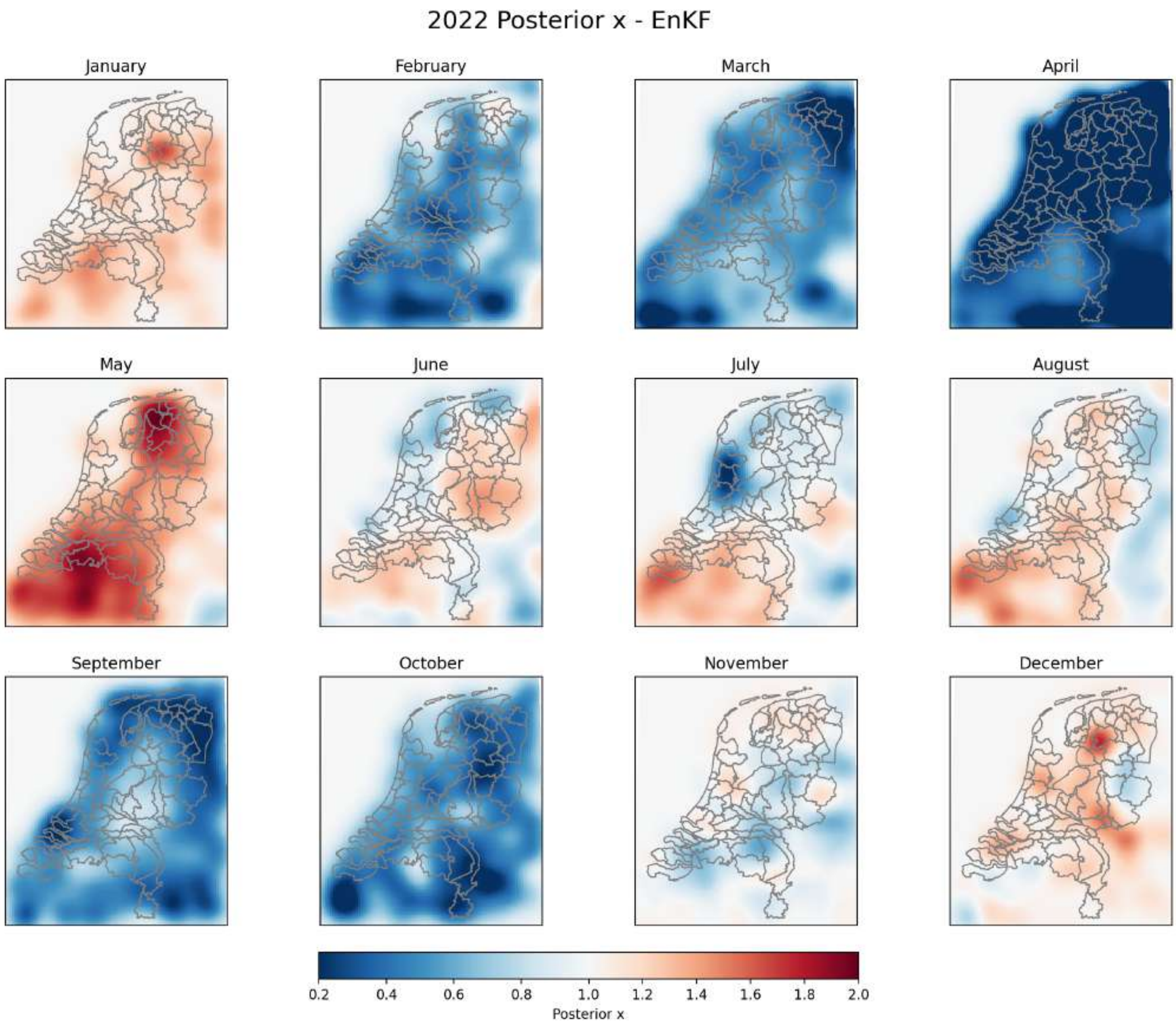


Figure S18. Monthly posterior emission scaling factors for 2022 using real IASI satellite only.

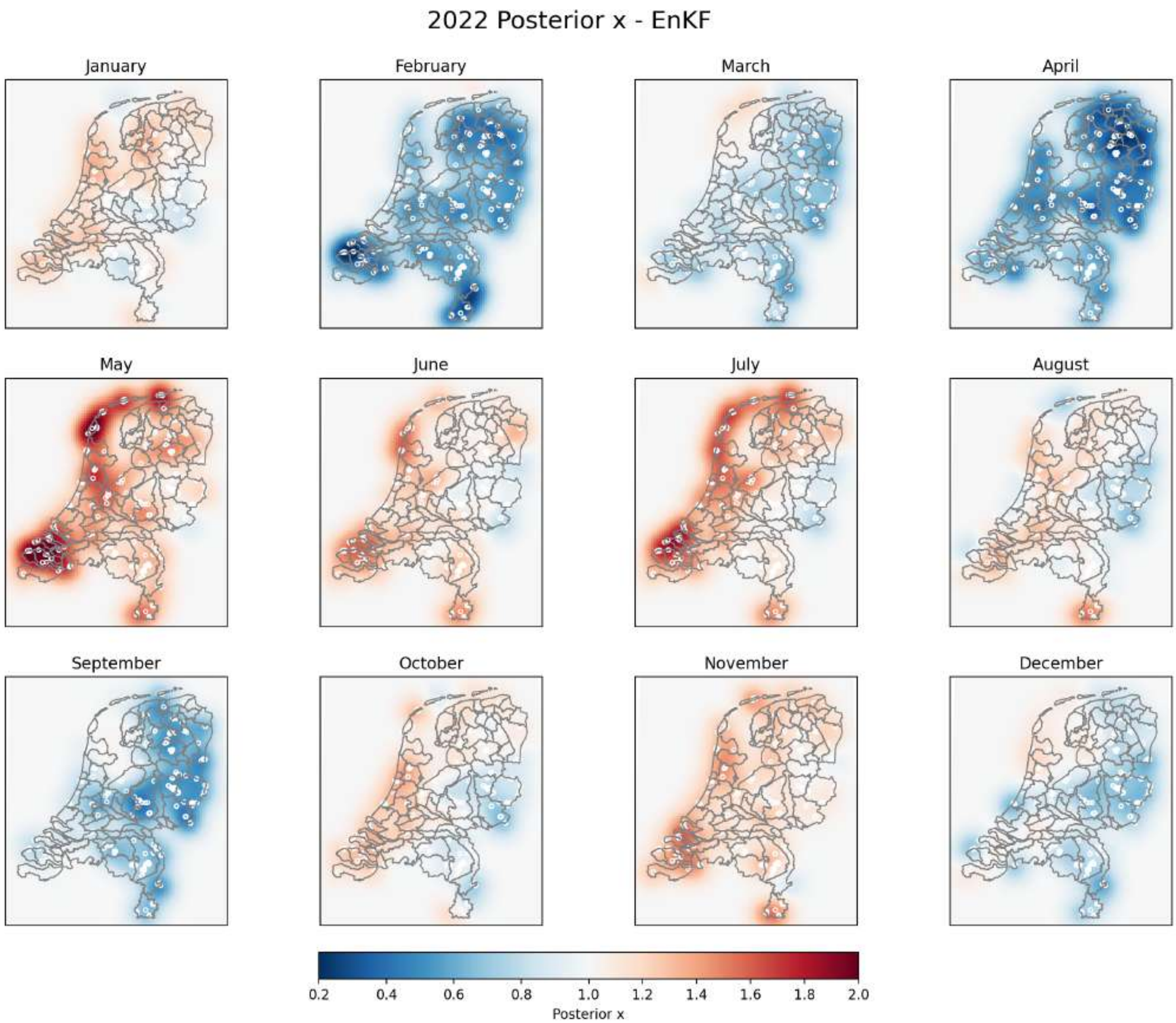


Figure S19. Monthly posterior emission scaling factors for 2022 using in situ observations (MAN+LML).

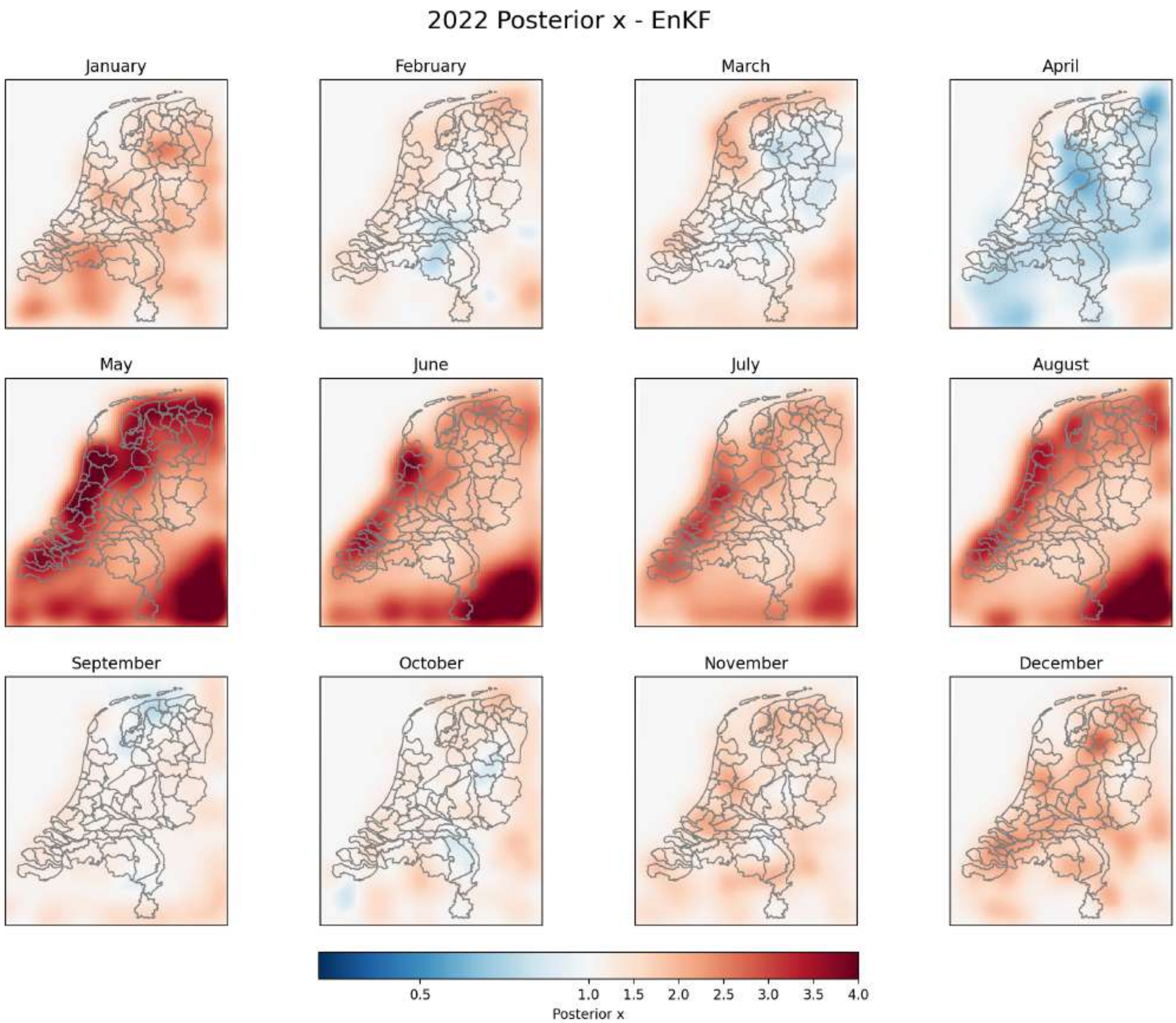


Figure S20. Monthly posterior emission scaling factors for 2022 using combined CrIS and IASI satellite observations.

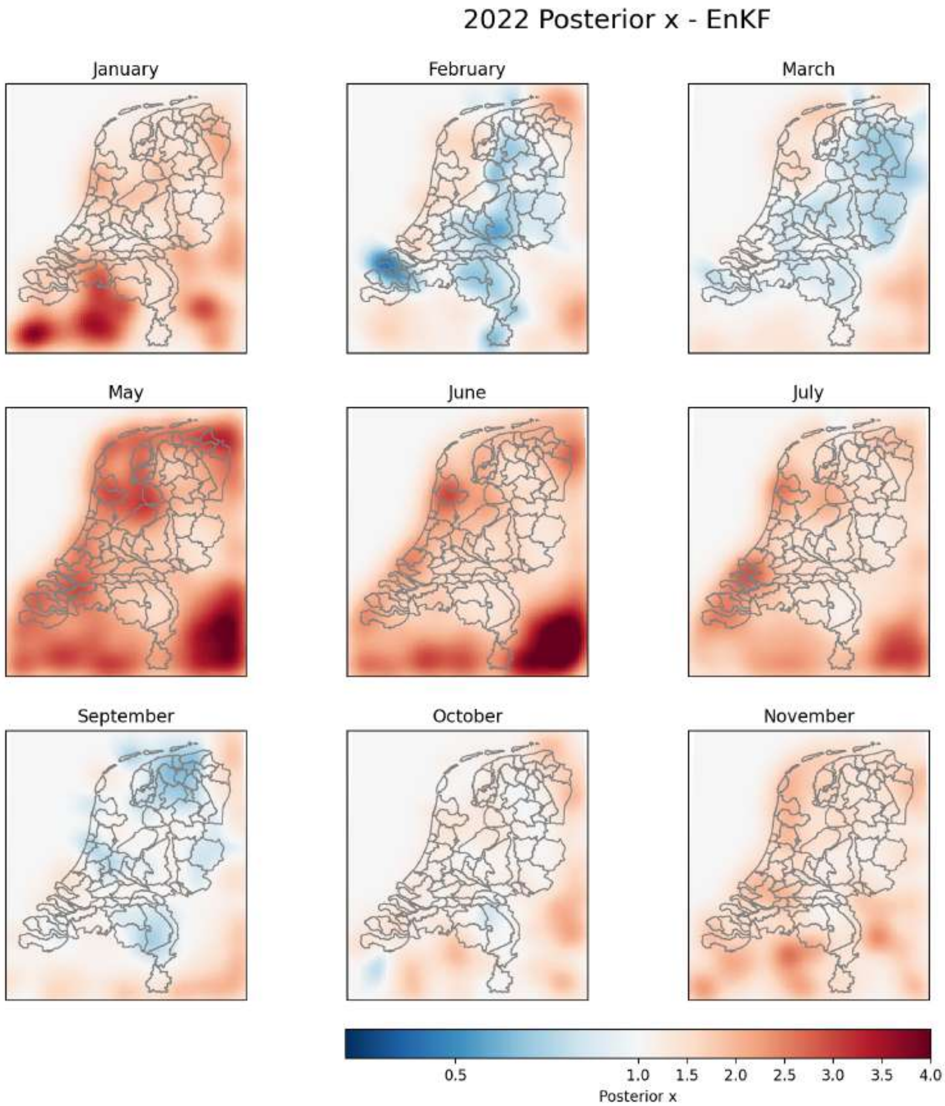


Figure S21. Monthly posterior emission scaling factors for 2022 using combined insitu (MAN+LML) and satellite (CrIS +IASI) observations.

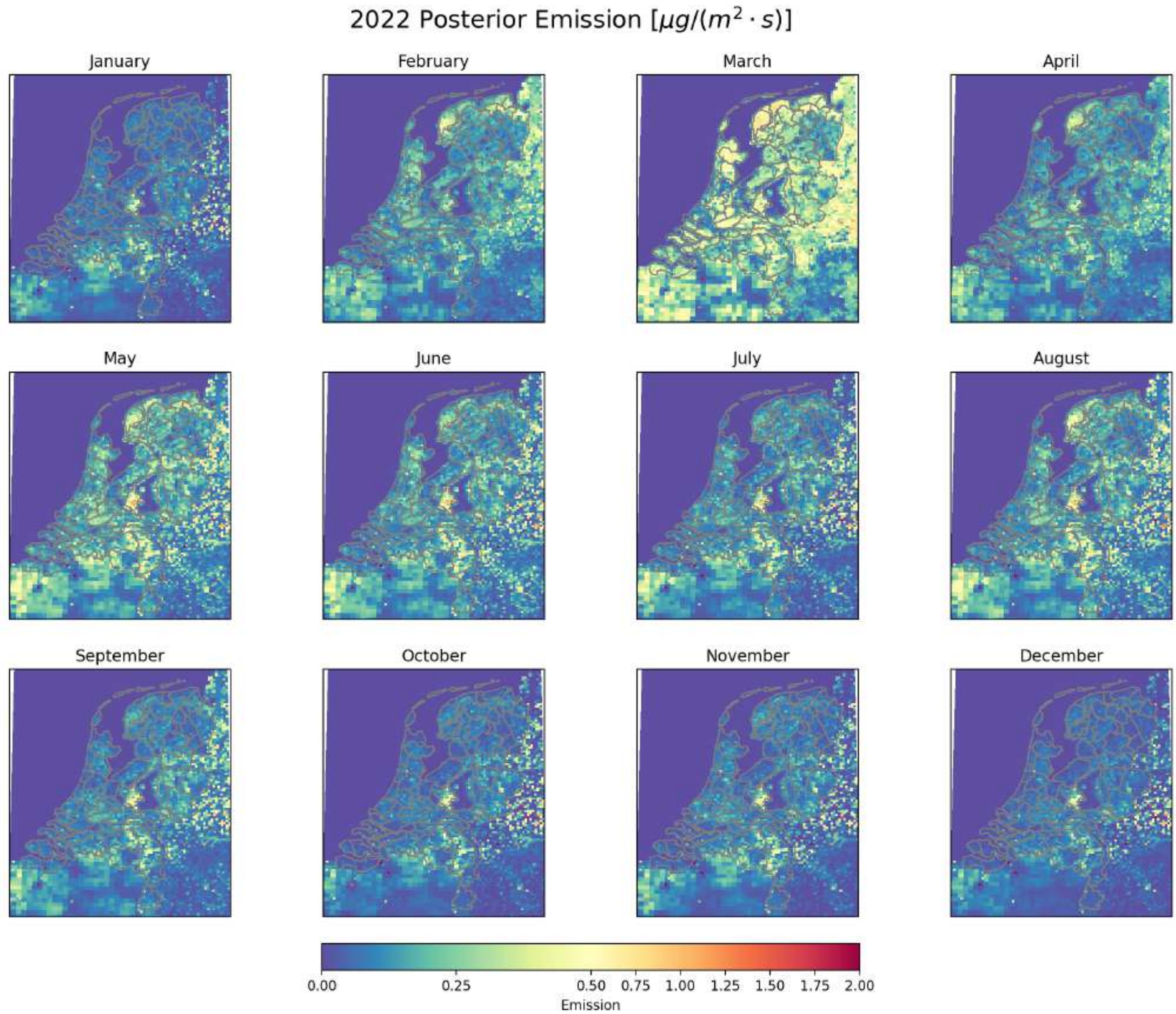


Figure S22. Monthly posterior emission for 2022 using combined insitu (MAN+LML) and satellite (CrIS +IASI) observations.

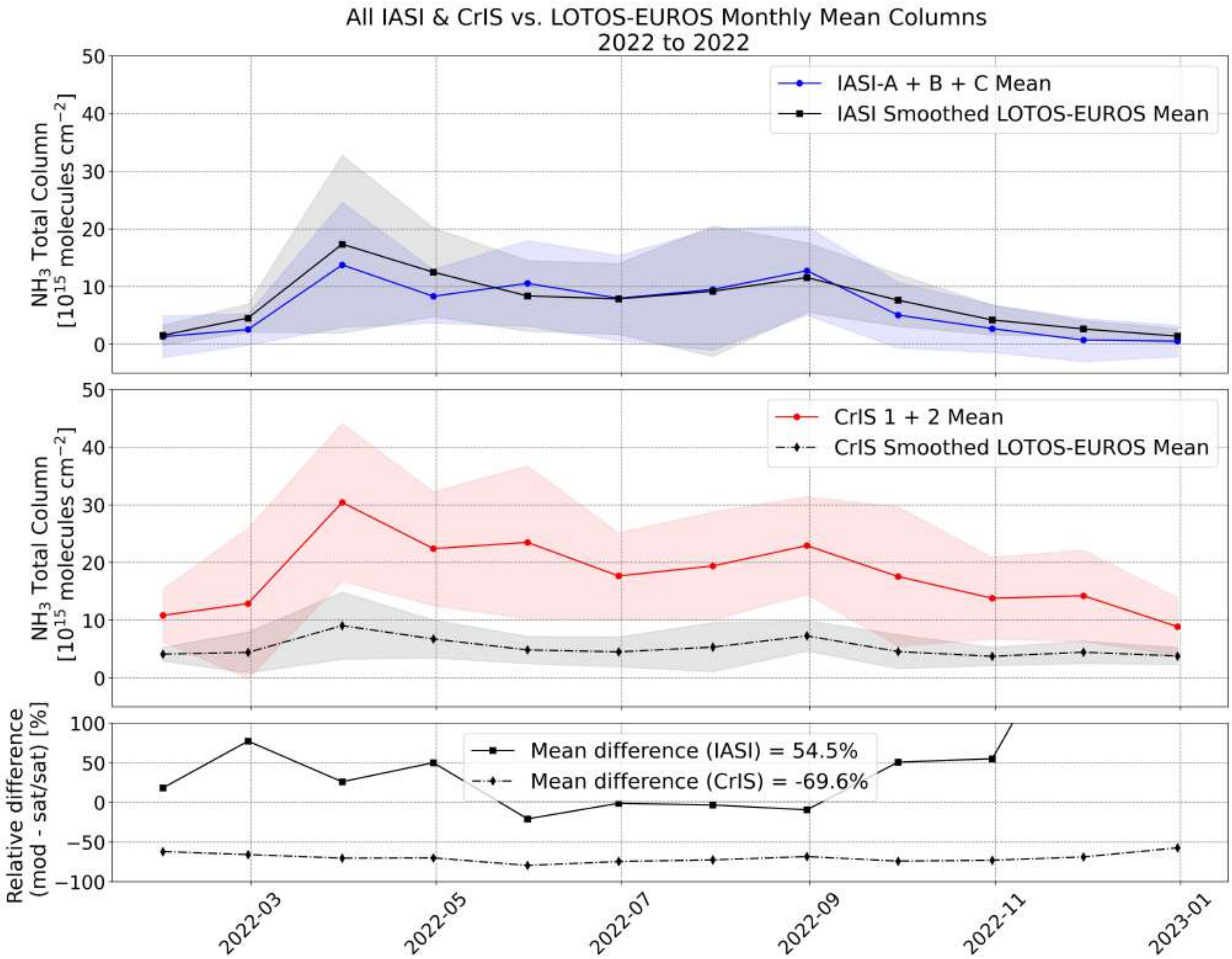


Figure S23. Monthly time profile of the retrieved satellite data in 2022.

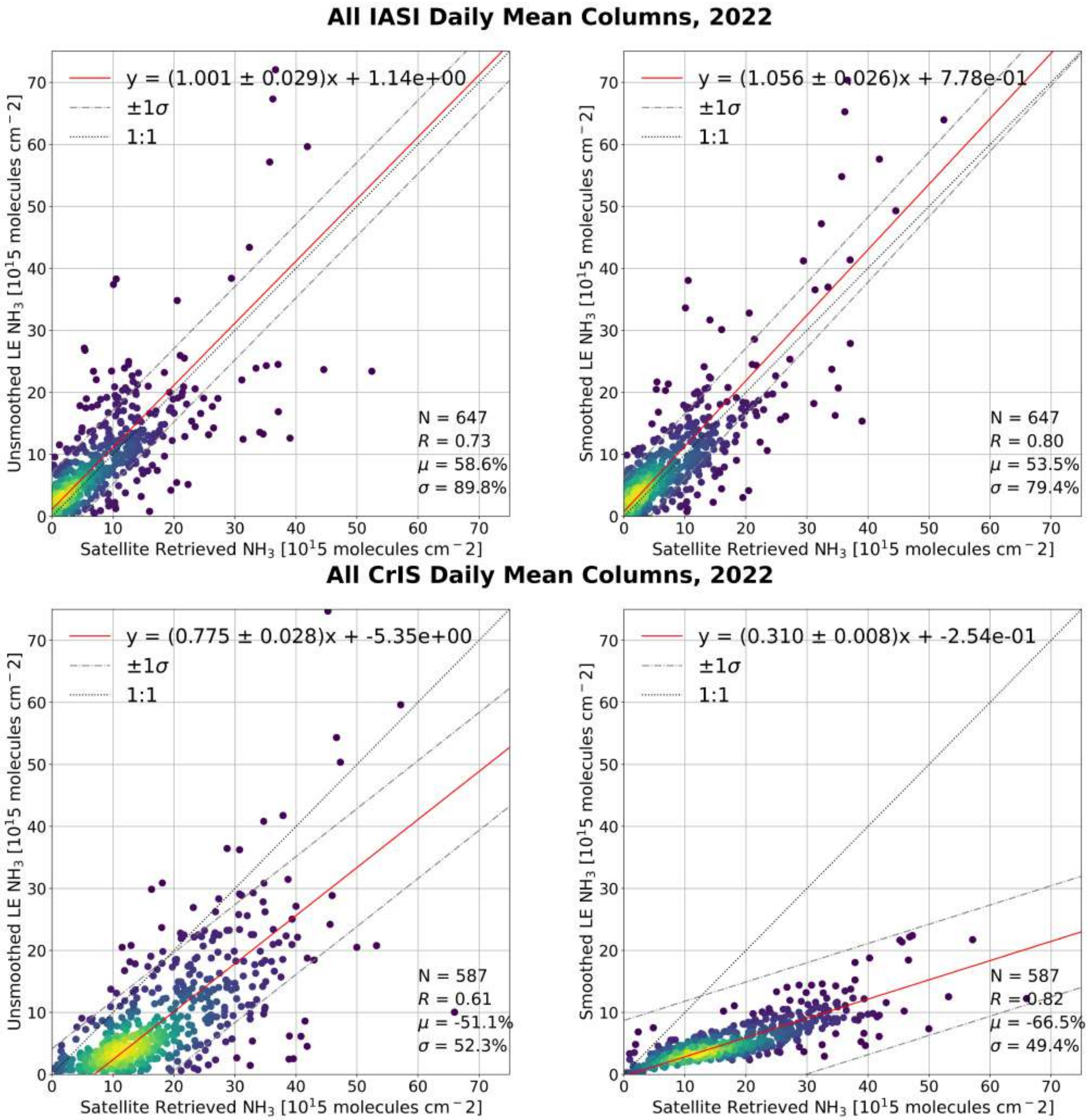


Figure S24. Scatter plot of simulated data and satellite retrievals.