Supplementary Materials: Combining The Emission Preprocessor HERMES With The Chemical Transport Model TM5-MP

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1. Flowcharts

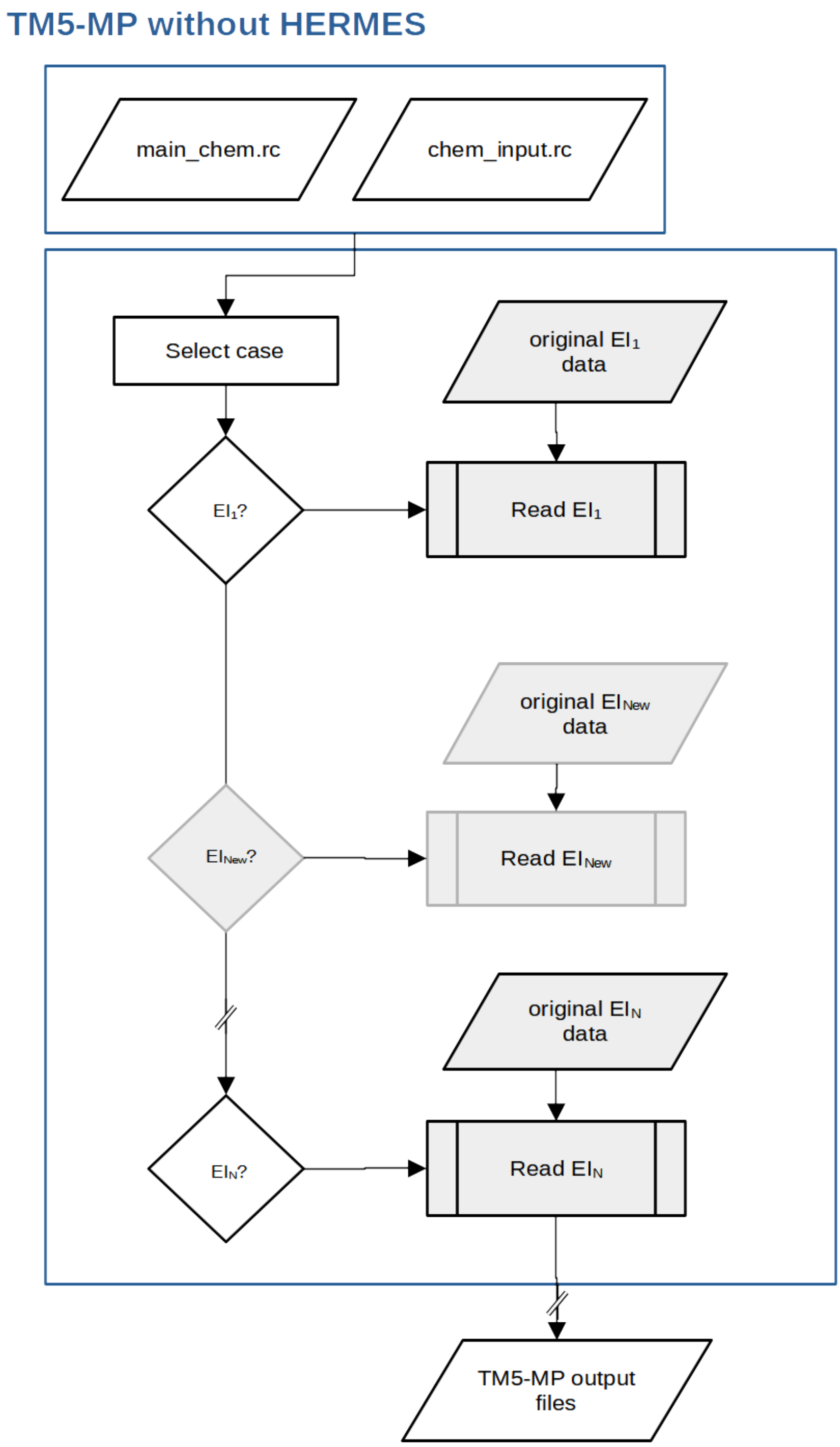

Figure S1. Flowchart for TM5-MP.

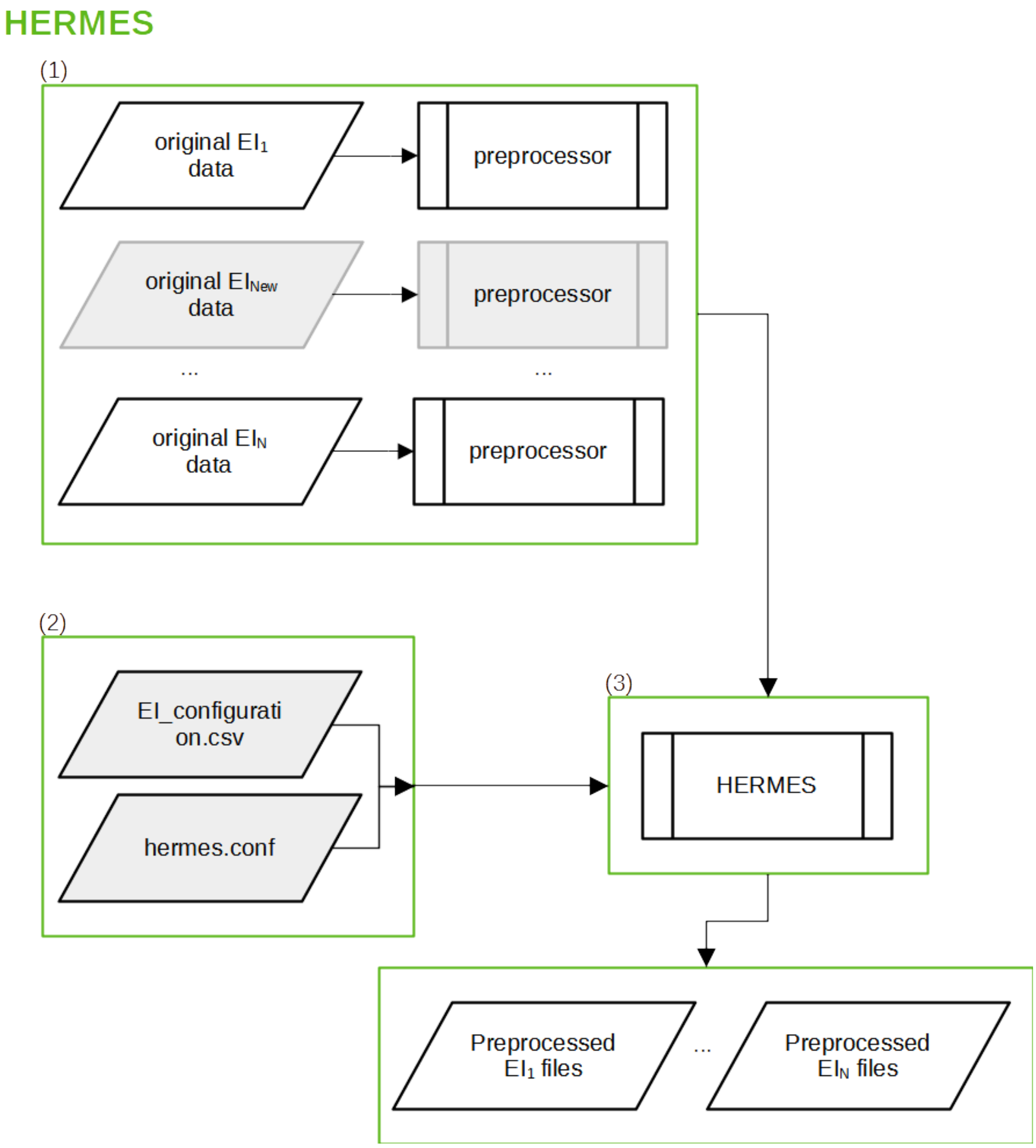

Figure S2. Flowchart for HERMES.

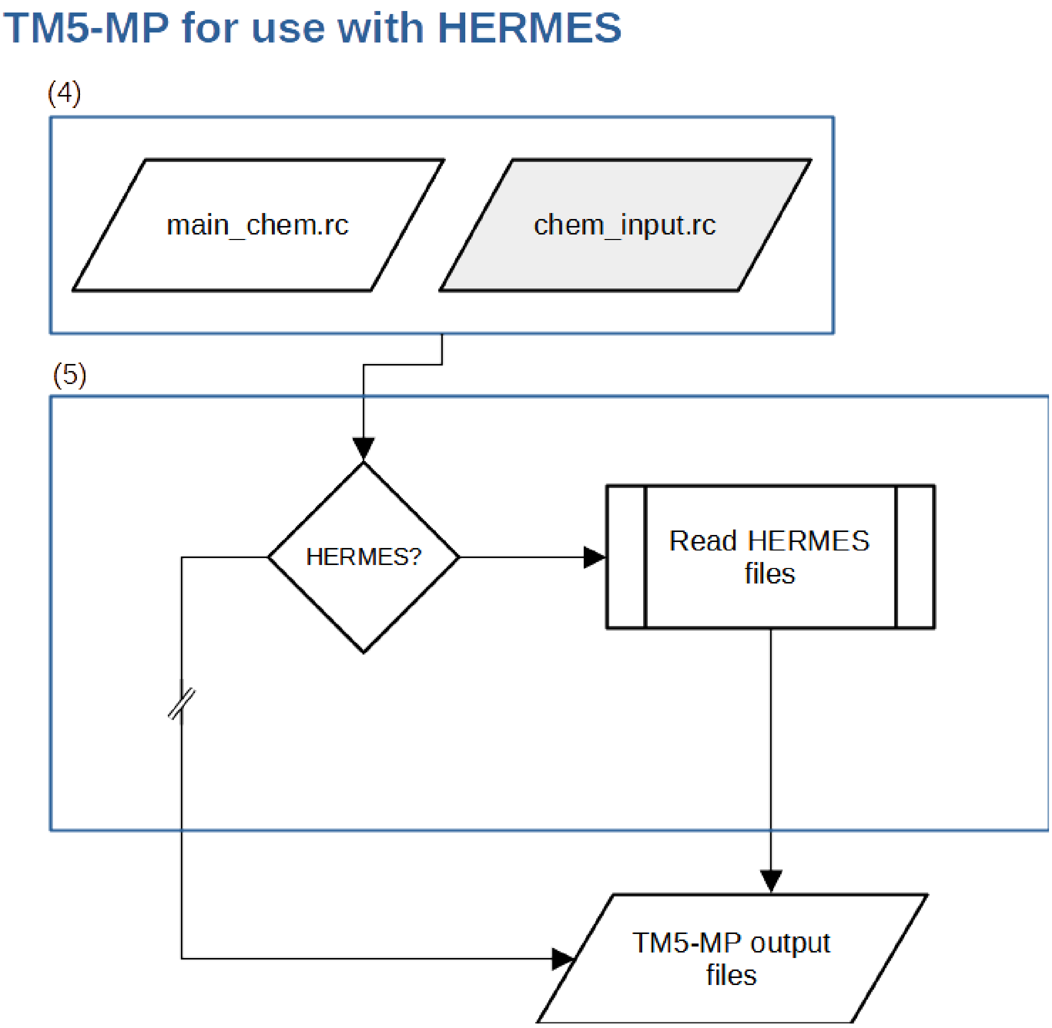

Figure S3. Flowchart for TM5-MP with new workflow.

2. Used Cluster

The model runs were performed on the HPC cluster Aether at the University of Bremen, financed by DFG within the scope of the Excellence Initiative. With 56 compute nodes: 2x Intel Xeon E5-2690v4 CPUs (2.6 GHz, 14 cores each, 16x 256 GB RAM, 40x 128 GB RAM)

3. Comparison of the old and new workflow

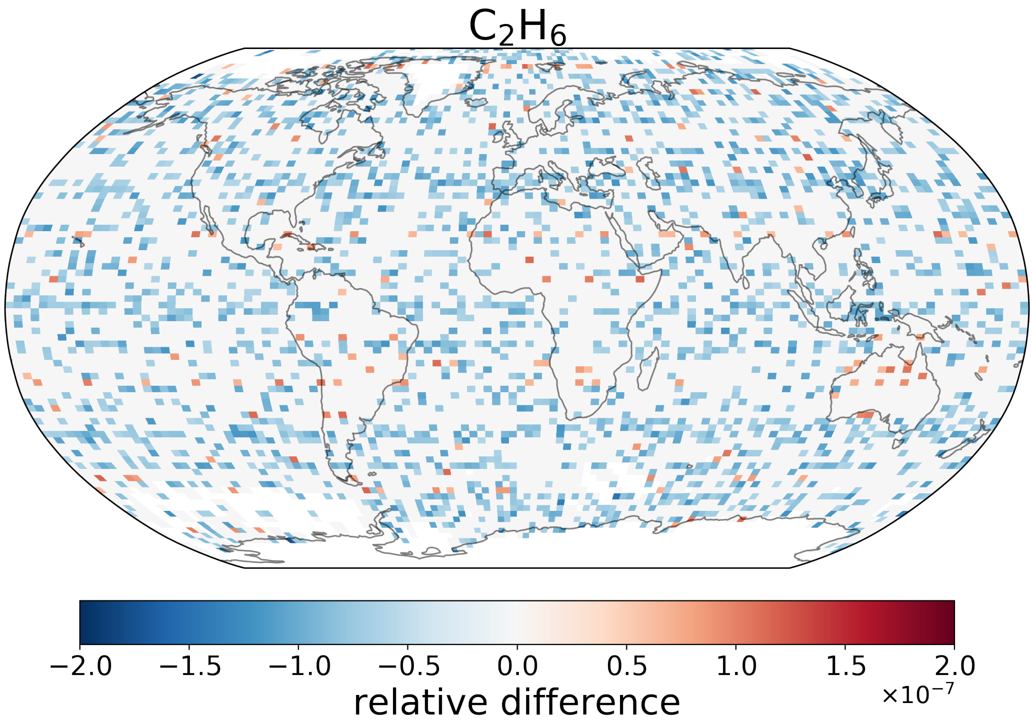

Figure S4. Differences between the resulting C_2H_6 budgets using the 'original' workflow (without the use of HERMES) and 'HERMES' workflow (with emission prepared through HERMES) relative to the results of the 'original' workflow.

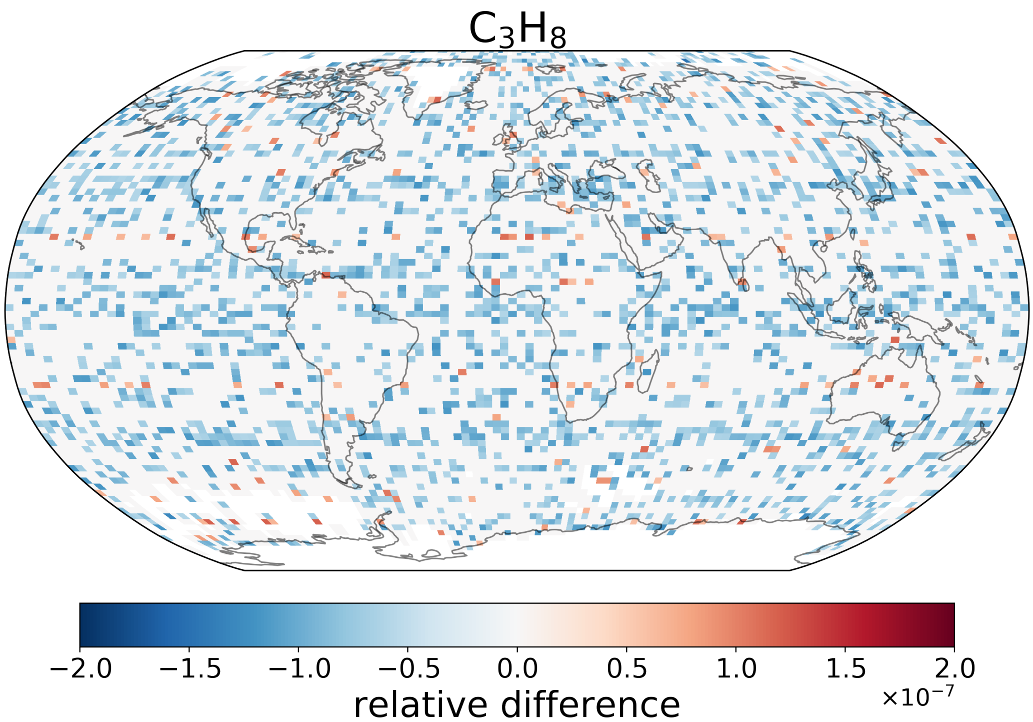

Figure S5. Differences between the resulting C_3H_8 budgets using the 'original' workflow (without the use of HERMES) and 'HERMES' workflow (with emission prepared through HERMES) relative to the results of the 'original' workflow.

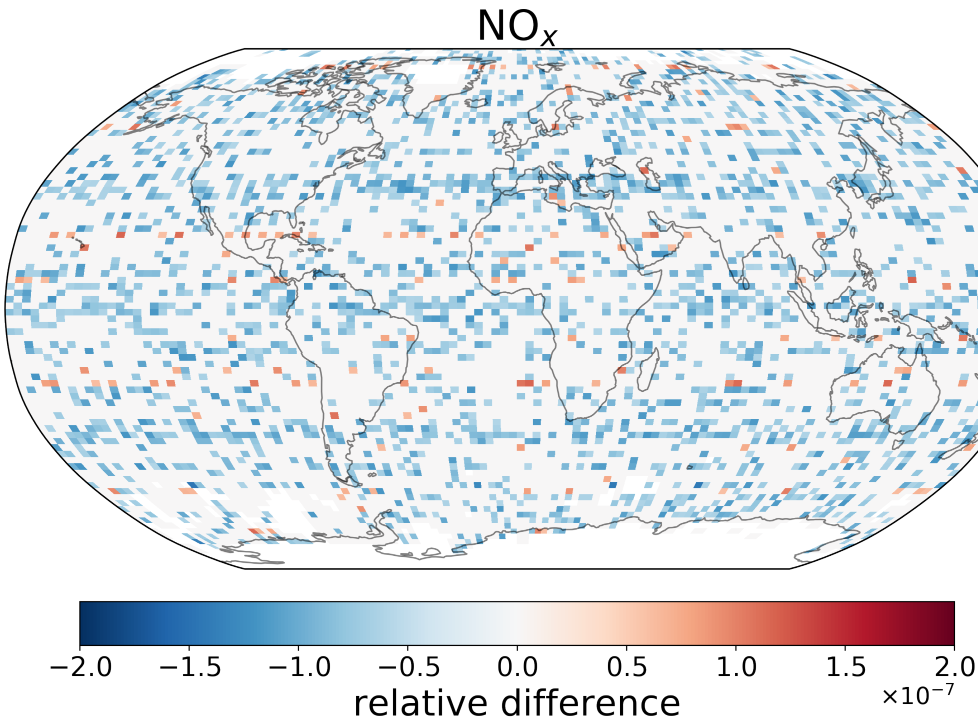

Figure S6. Differences between the resulting NO_x budgets using the 'original' workflow (without the use of HERMES) and 'HERMES' workflow (with emission prepared through HERMES) relative to the results of the 'original' workflow.

4. Measurements

4.1. Source of measurement data

Table S1. Sources of measurements used

Data source	Country / Region	Download Links	Last accessed
CNEMC (China National Environmental Monitoring Centre)	Mainland China	https://quotsoft.net/air/	15.05.2023
EPD (Environmental Protection Department), Hong Kong	Hong Kong	https://cd.epic.epd.gov.hk/EPICDI/air/station/?lang=en	27.04.2023
Ministry of Environment,Taiwan	Taiwan	https://data.moenv.gov.tw/dataset/detail/AQX_P_15	29.04.2023
National Institute of Environmental Sciences (NIES), Korea	Korea	https://www.airkorea.or.kr/web/last_amb_hour_data?pMENU_NO=123	24.04.2023
AAQMS (Ambient Air Quality Monitoring Stations) and RsAQMS (Roadside Air Quality Monitoring Stations), Japan	Japan	https://tenbou.nies.go.jp/download	08.05.2023
Nation Ambient Air Quality Monitoring Program (NAMP), Central Pollution Control Board (CPCB), India	India	https://app.cpcbccr.com/ccr/#/caaqm-dashboard-all/caaqm-landing	07.08.2023
AURN (Automatic Urban and Rural Network)	U.K.	directly downloaded by using the importAURN function in the openair R package	04.07.2023
AQS (Air Quality System), U.S.	U.S.	https://aqs.epa.gov/aqsweb/airdata/download_files.html	07.05.2023
National Air Pollution Surveillance (NAPS) program, Environment and Climate Change Canada	Canada	https://donnees-data.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program	22.06.2023
SINCA (Sistema de Información Nacional de Calidad del Aire), Chile	Chile	https://sinca.mma.gob.cl/index.php	27.09.2023
City of Cape Town, South Africa	Cape Town, South Africa	https://odp-cctegis.opendata.arcgis.com/documents/cctegis::air-quality/about	01.06.2023
Australian Capital Territory Government	Australian Capital Territory, Australia	https://www.data.act.gov.au/Environment/Air-Quality-Monitoring-Data/94a5-zqnn	06.06.2023
New South Wales Air Quality Monitoring Network	New South Wales, Australia	https://www.dpie.nsw.gov.au/air-quality/air-quality-data-services/data-download-facility	06.06.2023
Northern Territory Environment Protection Authority	Northern Territory, Australia	http://ntepa.webhop.net/NTEPA/Default.ltr.aspx	01.06.2023
Queensland Government	Queensland, Australia	https://www.data.qld.gov.au/dataset/air-quality-monitoring-2013 https://www.data.qld.gov.au/dataset/air-quality-monitoring-2014 https://www.data.qld.gov.au/dataset/air-quality-monitoring-2015 https://www.data.qld.gov.au/dataset/air-quality-monitoring-2016 https://www.data.qld.gov.au/dataset/air-quality-monitoring-2017	02.06.2023
South Australian Environment Protection Authority	South Australia, Australia	https://data.sa.gov.au/data/dataset/?tags=air+pollution	08.06.2023
Environment Protection Authority Victoria	Victoria, Australia	https://discover.data.vic.gov.au/dataset/epa-air-watch-all-sites-air-quality-hourly-averages-yearly/historical	11.06.2023
EANET (Acid Deposition Monitoring Network in East Asia)	East Asia (Mongolia), Southeast Asia (Indonesia, Thailand, Philippines)	https://monitoring.eanet.asia/document/public/index	15.03.2023
TOAR (Tropospheric Ozone Assessment Report) *	West Asia (Turkey), Europe (U.K. not included), Latin America (Mexico, Brazil, Colombia)	https://toar-data.org/surface-data	26.04.2023
National Oceanic and Atmospheric Administration (NOAA)	Global	https://gml.noaa.gov/aftp/data/trace_gases/co/flask/ https://gml.noaa.gov/aftp/data/trace_gases/voc/c2h6/flask/ https://gml.noaa.gov/aftp/data/trace_gases/voc/c3h8/flask/	24.07.2023 01.08.2023 01.08.2023

* Original sources of TOAR data in Europe include: (1) European Environment Agency (EEA)'s Airbase; (2) European Monitoring and Evaluation Programme (EMEP); (3) German Environment Agency's Air Data (UBA); (4) Institute of Geosciences, Dept. Meteorology, University of Bonn, Germany (MIUB). This information is found in: https://toar-data.fz-juelich.de/sphinx/TOAR_TG_Vol02_Data_Processing/build/html/data-sources.html

4.2. Location of measurement stations

For this study a large number of measurements were used to compare to the modelled mixing ratios. The location of these stations, as well as the providers are listed below, depending on their species.

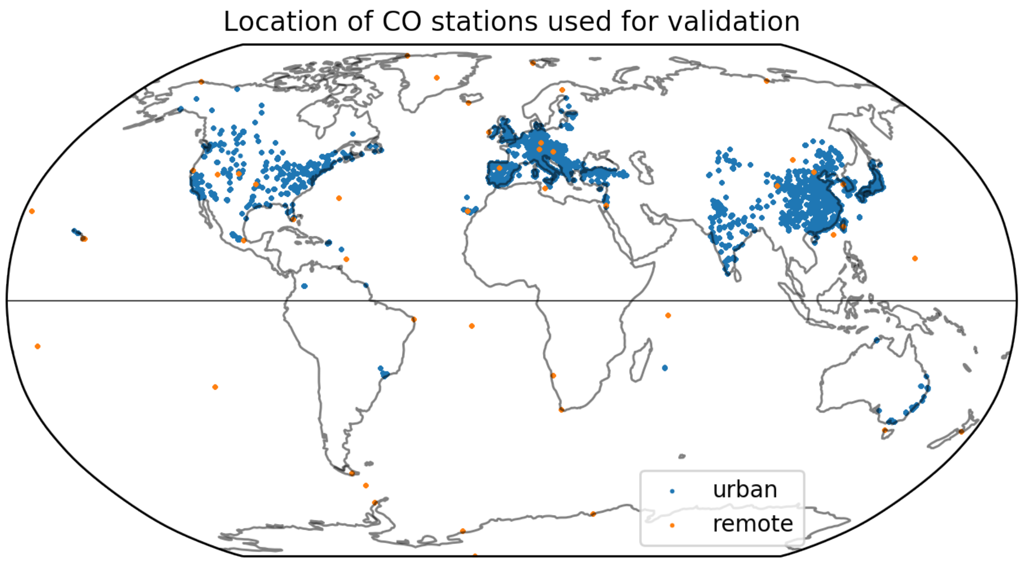

Figure S7. Location of CO measurement stations used for validation. Orange dots are representing NOAA stations ('remote'), while blue dots are showing stations closer to polluted regions.

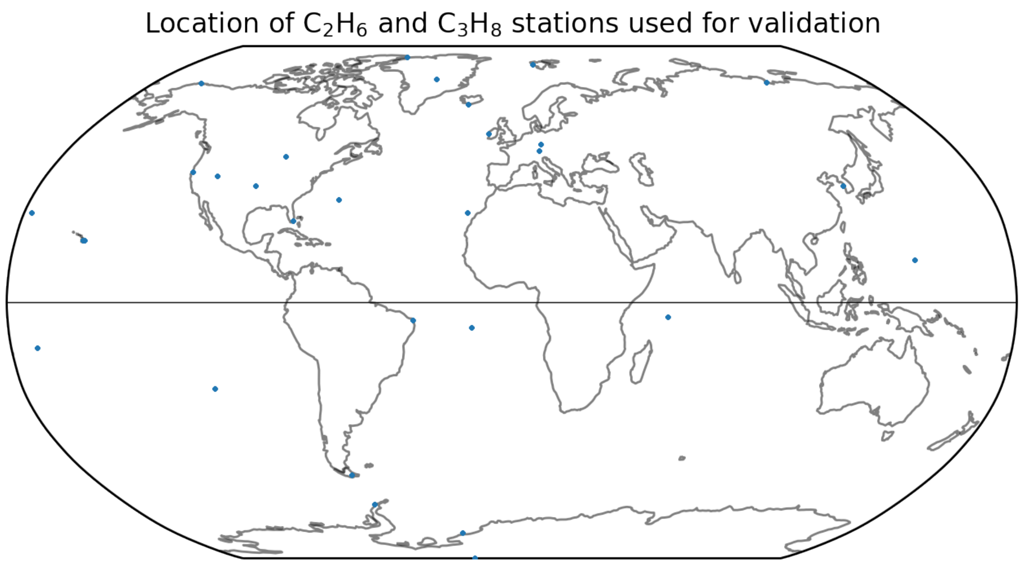

Figure S8. Location of C_2H_6 and C_3H_8 NOAA measurement stations used for validation.

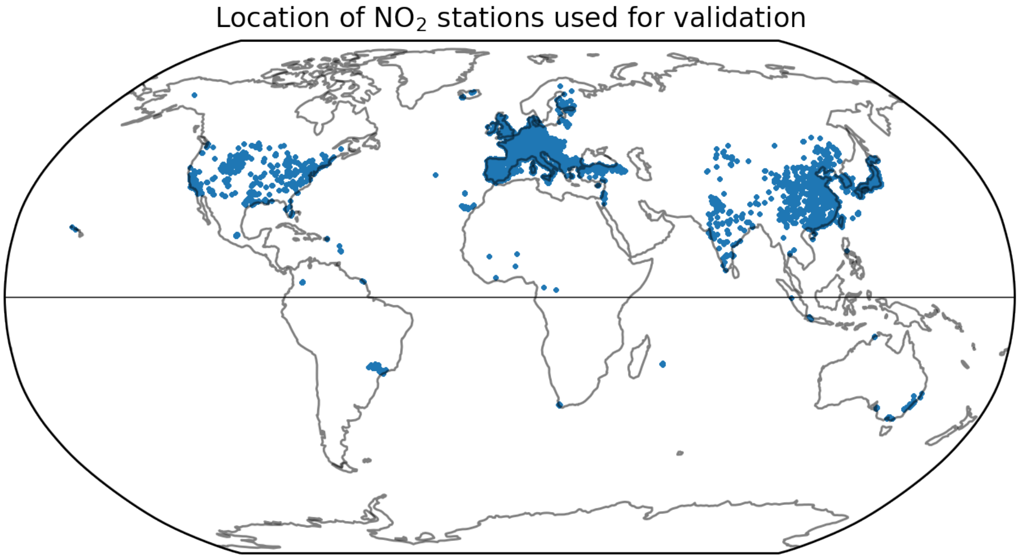

Figure S9. Location of stations used for validation with NO_2 measurement data.

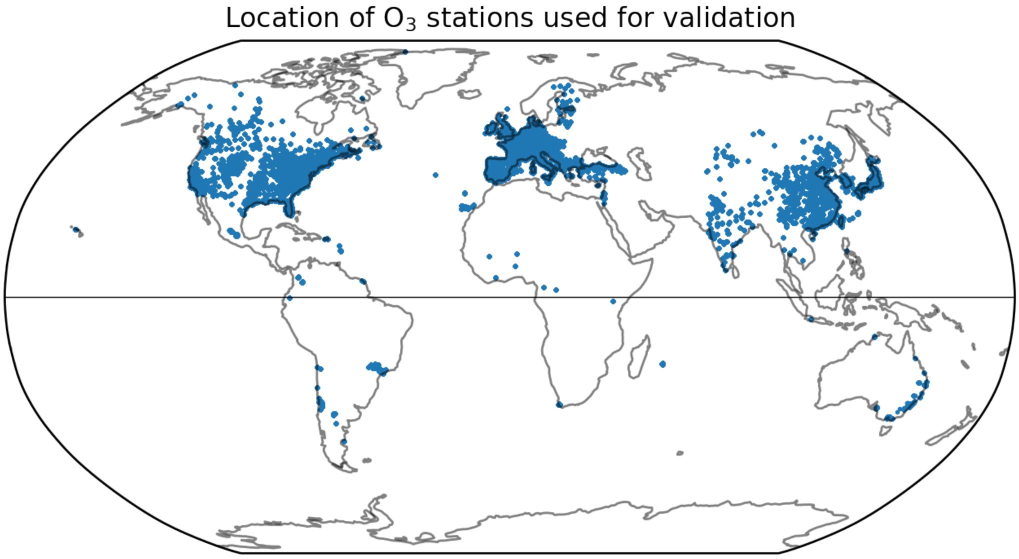

Figure S10. Location of stations used for validation with O_3 measurements.

5. Averaged difference between CEDS and REAS

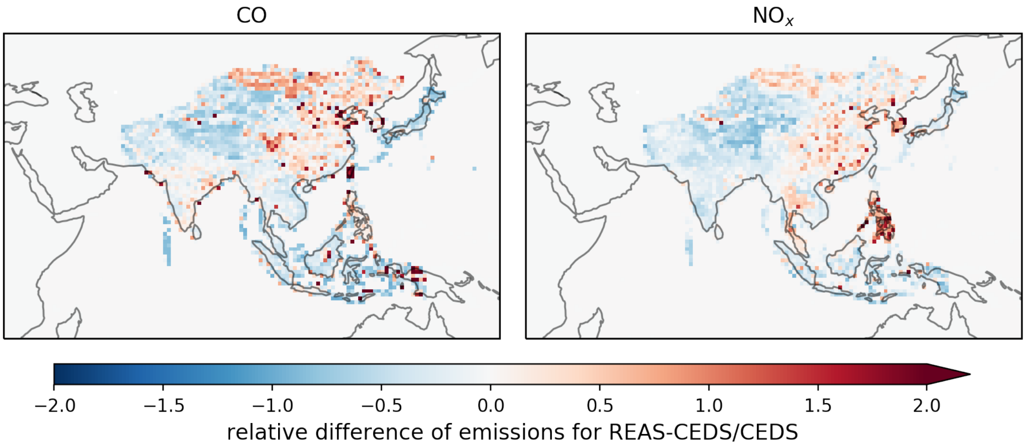

Figure S11. Relative difference between REAS and CEDS for CO and NO_2, averaged over the year 2012 – 2014.

Figure S11 shows the relative difference between REAS and CEDS for CO and NO_2. It can be seen, that for CO there are many regions having higher emissions in REAS seen as light red regions. This fits to the results seen from the validation, where an introduction of REAS data into the model is increasing the modeled mixing ratios. For NO_2, on the other hand, most of the region is displayed in blue, depicting smaller emissions in REAS. This also is inline with the results from the model validation, where the introduction of REAS leads to a reduction of NO_2 mixing ratios. One exception are the Philippines. However, no measurement data was available for this study, so the expected increase was not observed here.

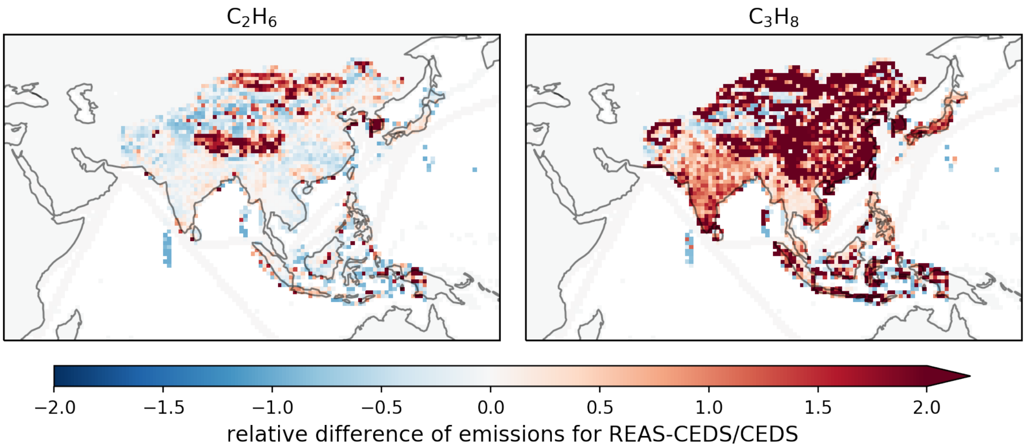

Figure S12. Relative difference between REAS and CEDS for C_2H_6 and C_3H_8, averaged over the year 2012 – 2014.

6. Results

6.1. Used metrics

To evaluate the differences between modeled results and measurements four different metrics were used. The bias and normalized mean bias were calculated based on the equations given below. The error where derived from the standard deviation of the differences between model and measurements, in case of the NMB additionally normalized to the measurements. The slope was derived from a fitted linear function. Furthermore, the Pearson correlation was calculated from the linear fit.

$$\mathrm{MB} = \frac{1}{N}\sum_{i=1}^{N}(M_i - O_i). \tag{1}$$

$$\mathrm{NMB} = \frac{1}{N}\frac{\sum_{i=1}^{N}(M_i - O_i)}{\sum_{i=1}^{N} O_i}. \tag{2}$$

6.2. Scatter plots

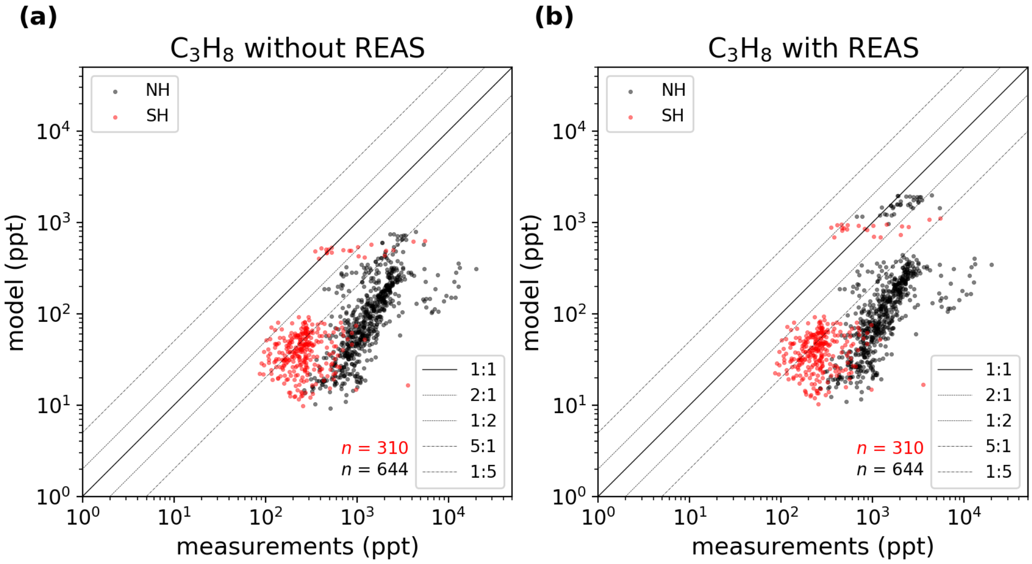

Figure S13. Comparison between the measured and modeled monthly-mean near-surface concentrations of ethane (C_3H_8). The results were split in stations in the Northern Hemisphere (NH, in black), and the Southern Hemisphere (SH, in red). The results in the simulation without and with REAS are shown in the panel a and b, respectively. The solid black line is the 1:1 line, while the dashed lines are the 1:5, 1:2, 2:1 and 5:1 lines.

Figure S13 shows the comparison of modeled and measured results of propane at several NOAA stations. The model results were analyzed for the model runs with and without REAS data included. The results for the Northern Hemisphere look similar to those for ethane shown in the main part of the manuscript. For the Southern Hemisphere (SH) one station shows a significant larger modeled propane than the rest of the stations. These values belong to a measurements station in Bukit Kototabang in Indonesia. The good agreement of modeled and measured propane at that station, which even increases with the use of REAS, results in a significant improvement of the averaged bias from all stations in the SH. However, this improvement is not as significant at the other stations, if existing at all.

6.3. Statistics

The statistics resulting from the comparison of modeled and measured mixing ratios for C_3H_8, NO_2 as well as O_3 are presented here.

Table S2. Statistics for the model performance of C_3H_8 in comparison to measurements at remote stations.

	without REAS		**with REAS**	
	NH	SH	NH	SH
MB (ppt)	-1543 ± 1672	-289 ± 432	-1464 ± 1676	-263 ± 407
NMB	-0.92 ± 1.00	-0.80 ± 1.20	-0.87 ± 1.00	-0.73 ± 1.13
slope	0.03	0.14	0.05	0.25
r^2	0.39	0.57	0.22	0.57

Table S3. Statistics for the model performance of NO_2 in comparison to measurements at urban stations.

	without REAS		**with REAS**	
	Asia	outside Asia	Asia	outside Asia
MB (ppb)	-10.0 ± 7.1	-5.9 ± 4.7	-10.3 ± 7.0	-5.9 ± 4.7
NMB	-0.59 ± 0.42	-0.68 ± 0.55	-0.61 ± 0.42	-0.68 ± 0.55
slope	0.33	0.19	0.34	0.19
r^2	0.56	0.44	0.58	0.44

Table S4. Statistics for the model performance of O_3 in comparison to measurements at urban stations.

	without REAS		**with REAS**	
	Asia	outside Asia	Asia	outside Asia
MB (ppb)	15 ± 12	14 ± 7	17 ± 12	13 ± 7
NMB	0.54 ± 0.43	0.50 ± 0.27	0.59 ± 0.41	0.48 ± 0.27
slope	0.80	0.57	0.80	0.56
r^2	0.60	0.62	0.61	0.61