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Dalsøren, Stig B., Myhre, Gunnar, Hodnebrog, Øivind et al. (14 more authors) (2018) Discrepancy between simulated and observed ethane and propane levels explained by underestimated fossil emissions. *Nature Geoscience*. pp. 178-184. ISSN 1752-0908

<https://doi.org/10.1038/s41561-018-0073-0>

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1 **Discrepancy between simulated and observed ethane and propane levels explained by**
2 **underestimated fossil emissions**

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27 **Ethane and propane are the most abundant non-methane hydrocarbons in the atmosphere.**
28 **However, their emissions, distribution in the atmosphere, and trends in their atmospheric**
29 **concentrations are insufficiently understood. Atmospheric model simulations using**
30 **standard community emission inventories do not reproduce available measurements in the**
31 **Northern Hemisphere. Here, we show that observations of preindustrial and present-day**
32 **ethane and propane can be reproduced in simulations with a detailed atmospheric-**
33 **chemistry transport model, provided natural geologic emissions are taken into account and**
34 **anthropogenic fossil fuel emissions are assumed to be two to three times higher than**
35 **indicated in current inventories. Accounting for these enhanced ethane and propane**
36 **emissions results in simulated surface ozone concentrations that are 5 to 13 % higher than**
37 **previously assumed in some polluted regions in Asia. The improved correspondence with**
38 **observed ethane and propane in model simulations with greater emissions suggests that the**
39 **level of fossil (geologic + fossil fuel) methane emissions in current inventories may need re-**
40 **evaluation.**

41

42 Direct emissions at the surface are the only sources of ethane and propane to the atmosphere^{1,2},
43 and several studies suggest that they are underestimated in global inventories^{1,3-7}. A major source
44 of uncertainty is that these inventories first calculate total non-methane hydrocarbon (NMHC)
45 emissions and then disaggregate them into individual species (ethane, propane, etc.) based on
46 limited amounts of data⁸⁻¹¹. Over the past decade the inventories do not fully account for an
47 abrupt increase in the exploitation of unconventional natural gas in the United States^{3,4} and
48 therefore likely underestimate present-day emissions^{3,4,12-23}. Recent atmospheric model
49 simulations applying current global emission inventories tend to underestimate observed ethane
50 and propane concentrations in wintertime in the Northern Hemisphere^{3-5,7,12,24-29}.

51 Anthropogenic fossil fuel (conventional and unconventional) emissions are presently the largest
52 emission source of ethane and propane in most global inventories. Fugitive emission is the main
53 fossil fuel NMHC source and includes venting and flaring, evaporative losses, and equipment
54 leaks but not fuel combustion. Changes in these particular emissions are regarded as the main
55 cause of observed ethane trends^{3,4,26,30-34}. Recent studies^{9,35} calculated fugitive fossil fuel
56 emissions from oil, natural gas, and coal systems for ethane based on a joint inventory and

57 atmospheric box-model approach. Their emission dataset⁹ was recently updated with new data³⁶.
58 Another recent study¹⁰ used a detailed inventory approach to identify cold venting of associated
59 petroleum gas containing also methane, propane, and butane as significant emission sources
60 potentially underestimated in existing emission inventories. These new studies combine field
61 measurements and country-specific information from published sources along with observed
62 flaring of associated gas from satellite images. In combination these take into account that the
63 emission factors from venting and flaring of associated gas released during extraction vary
64 considerably across different oil, coal, and gas fields around the world. Such considerations have
65 not been made in most community emission datasets, which apply emission factors reported by
66 countries to the United Nations Framework Convention on Climate Change, or from
67 measurements representative for North America for those countries that are not reporting.
68 Deficiencies in fugitive fossil fuel emission estimates in community emission datasets were also
69 recently found for black carbon at high latitudes³⁷ and SO₂ at low latitudes³⁸ (the Middle East).
70 Natural geologic emissions are another suggested fossil hydrocarbon source missing in
71 inventories^{39,40}, receiving little attention in previous model studies. Major geologic sources
72 include seepage from onshore and submarine petroleum basins, volcanoes, and degassing from
73 geothermal manifestations (see Methods). Based on the few available estimates^{39,40}, geologic
74 emissions may have been the largest preindustrial source of ethane to the atmosphere (Figure
75 1a).

76 Another cause of poor model performance could be inaccurate representation of atmospheric
77 sinks. Oxidation by hydroxyl in the troposphere is the main sink for ethane and propane^{1,2,41}.
78 Estimates of global mean hydroxyl levels and hemispheric ratios differ substantially between
79 observation-based estimates and results from model ensembles⁴²⁻⁴⁴. Studies also find large inter-
80 model differences in the atmospheric distributions of oxidants⁴⁵.

81 In this study we first apply the OsloCTM3 model⁴⁶ to investigate the preindustrial atmospheric
82 ethane budget. We compare model results from simulations with and without geological
83 emissions to ice-core measurements. Thereafter, we model current conditions represented by the
84 year 2011, which is the last year available in all applied fugitive fossil fuel emission datasets. In
85 the year 2011 baseline simulation we use the state of the art global anthropogenic emission
86 inventory CEDS CMIP⁴⁷. We also include natural emissions (treated as negligible in many

87 model studies) from oceanic, biogenic (vegetation) and geologic sources (see Methods). We
88 compare with surface ethane and propane measurements from global and regional surface
89 networks (see Methods) with a focus on observations in the mid- to high-latitude Northern
90 Hemisphere where previous model studies underestimated observations. We then generate
91 alternative gridded emissions by replacing the fugitive fossil fuel emissions in the CEDS CMIP6
92 inventory with new datasets that better represent fossil fuel activity and emissions^{9,10,35,36} with
93 corrections to avoid double counting from potential overlap with natural geologic emissions (see
94 Methods). In the resulting alternative simulations (ALT1 and ALT2, Tables 1-2) the fossil fuel
95 emissions are factors of about two (ethane, Figure 1b) and three (propane, Figure S1) higher than
96 in the baseline simulation with very different geographical distributions (Figure S2). We also
97 suggest further modifications of the ALT1 and ALT2 emissions based on studies with the
98 Flexpart model⁴⁸. Back-trajectories from Flexpart are used to identify source regions related to
99 OsloCTM3 under- and overestimation of observed ethane concentrations (see Methods). Finally,
100 we explore the uncertainty of the atmospheric sinks in a sensitivity study where we perturb the
101 OsloCTM3 hydroxyl level within its uncertainty range (see Methods).

102

103 **Preindustrial ethane budget**

104 Ice core measurements³⁹ reveal a large, factor of about 4, north/south inter-polar ratio for ethane.
105 Figure 2a shows that this can be reproduced by the model with a geologic source of 3 Tg/yr,
106 constituting about 40% of the total preindustrial ethane emissions (Figure 1a). With geologic
107 emissions included in the simulations the modeled abundance at Summit in Greenland agrees
108 with observations, and the simulated inter-polar ratio improves substantially relative to
109 observations. Without geologic sources the simulated abundance at Summit is 50 % too low.
110 This is in agreement with the findings of ref. 39, where a simpler model without interactive
111 oxidation chemistry was used.

112 Our baseline simulation slightly overestimates the observed Antarctic ethane concentration.
113 Transport to high southern latitudes and biomass burning emissions have high inter-annual
114 variability. However, sensitivity simulations with meteorological input data for a different year
115 and an alternative inventory with different geographical distribution and emission totals for
116 biomass burning emissions (see Figure 1a and Methods) resulted in minor changes (see

117 Supplementary). We therefore suggest that a small Antarctic overestimation relates to
118 uncertainties in the magnitude and distribution of geologic and oceanic emissions in the Southern
119 Hemisphere.

120

121 **Current ethane and propane budgets**

122 The baseline simulation for 2011 does not reproduce the observed inter-polar ethane ratio well,
123 even if geologic emissions are included (Figure 2b). The modeled concentration at Summit in
124 Greenland is only about 50 % of measured values (Figure 2b). Underestimations of ethane and
125 propane concentrations at high northern latitudes, particularly during wintertime (Figure 3,
126 Figures S3-S7), are similar to most other model studies^{3-5,7,12,24-29} using standard emission
127 inventories. As shown below, the likely cause is underestimated fossil fuel emissions in the
128 standard community emission data set CEDS CMIP6, used in the baseline simulation. The CEDS
129 CMIP6 emission data⁴⁷ agree with previous emission data for ethane and propane used in
130 atmospheric climate and air pollution studies (Figure 1b and Figure S1).

131 The ALT1 and ALT2 simulations, where the CEDS CMIP6 fugitive fossil fuel emissions are
132 replaced with the new datasets^{9,10,35,36}, reproduce the inter-polar ethane ratio and the observed
133 levels in Greenland (Figure 2b), Zeppelin Observatory at Svalbard (Figure 3a), and most other
134 stations (Figure 3b, Figures S3-S9) much more closely. This is also the case for propane (Figure
135 3a, c, Figures S3-S9), for which fossil fuel emissions play an even larger role (Figure S1). A
136 substantial improvement is found throughout the Arctic (Figure S5, Tables S2-S3). ALT1
137 performs better than ALT2. Both have positive mean biases, tending to overestimate episodes
138 with high concentrations. We explore these and other episodes at Zeppelin (Arctic station with
139 frequent sampling) in a systematic way (see Methods). Figure 4 shows that the episodes with the
140 largest underestimation of ethane at Zeppelin in the baseline simulation occur for air masses
141 originating from Eurasia. Fossil fuels are the dominant emission source in this region for most of
142 the year, strongly suggesting that these are underestimated in the CMIP6 inventory. From Figure
143 4, it is also evident that ALT1 underestimates the fossil fuel emissions in northwestern Europe
144 (i.e. mainly emissions from the North Sea) while it overestimates emissions from Russia. The
145 ALT2 simulation mainly overestimates observed ethane levels at Zeppelin (Figure 3a). For this
146 inventory, the fossil fuel emissions are likely overestimated both in the North Sea and Russia

147 (Figure 4). Overestimated emissions in ALT1 and ALT2 over Russia also seem likely from the
148 comparison with Tiksi station data (Figure S5, the only available station in Russia).

149 At mid-latitude stations in the U.S. and Canada, both ALT1 and ALT2 show good agreement
150 with measurements (Figure S6, Tables S2-S3). An exception is the highly oil and natural gas
151 influenced Southern Great Plains station (large underestimation) (Figure S6). Large emissions
152 from several nearby oil wells might not be fully resolved in the model averaging emissions over
153 the model grid scale. It is also a possibility that the ALT1 and ALT2 inventories underestimate
154 the emissions from nearby unconventional gas fields (e.g. Woodford, Barnett⁴⁹) and oil wells.
155 The ALT1 and ALT2 simulations also improve agreement with measurements at non-Arctic
156 European stations compared to the baseline (Figure S7, Tables S2-S3), especially for ethane. At
157 most stations the ALT1 simulation is biased slightly low compared to the observations. ALT2
158 also performs better than the baseline, but overestimates, to varying degree, the measurements at
159 most European stations.

160 The lower latitude Cape Verde site also shows large improvements (ALT simulations vs.
161 baseline) during wintertime (Figure S8) when it is influenced by air passing over the Sahara⁵⁰
162 from oil and gas fields in northern Africa and the Middle East. In the Southern Hemisphere, the
163 baseline simulation reproduces observed levels and seasonal patterns well and the alternative
164 simulations only result in minor differences (Figures 2-3 and Figure S9).

165 The ALT1 and ALT2 anthropogenic ethane emissions (excluding biomass burning) are slightly
166 smaller than the optimized anthropogenic emissions in other recent model studies^{3,4} (Figure 1b).
167 The optimized emissions in other studies are based on sensitivity simulations^{3,4} finding that an
168 approximate doubling of anthropogenic emissions is needed to reproduce measurements at
169 Jungfrauoch⁴ and a few other FTIR stations in the Northern Hemisphere³. In our study we
170 include natural geologic emissions and apply new detailed emission datasets for fugitive fossil
171 fuel emissions instead of performing an up-scaling of all anthropogenic emissions. Our model is
172 also run at higher spatial and temporal resolution and compared to a larger number of
173 measurement sites.

174 Overestimated atmospheric loss (i.e. too high hydroxyl levels) might lead to an underestimation
175 of observed ethane and propane levels. However, this cannot be a major cause for the
176 discrepancies in our baseline simulation. Scaling down tropospheric hydroxyl levels to the lower

177 range of model and observational based estimates (see Methods) improves the agreement slightly
178 (Figure S10) but much of the underestimation in the mid-high Northern Hemisphere during
179 wintertime persists. This seasonal pattern is expected as chemical loss at high northern latitudes
180 is inefficient during wintertime when little sunlight and low water vapor concentrations result in
181 low hydroxyl concentrations.

182 We have not included atmospheric oxidation of ethane and propane by halogens in the model
183 simulations. The reasons and implications on uncertainty are discussed in the Supplementary.
184 Inclusion of halogen chemistry would likely lead to slightly larger underestimation of ethane and
185 propane in the baseline simulation, thereby supporting that emissions in standard community
186 datasets are too low.

187

188 Our global total natural geologic emissions correspond with the best estimates from the study by
189 ref. 40. Reported uncertainty ranges^{39,40} are shown in Figure 1 and Figure S1. A recent study⁵¹
190 suggests geologic methane emissions about 1/3 of that estimated by ref. 40. NMHC to methane
191 emission ratios from geologic sources are uncertain and likely highly spatially and temporally
192 variable. Therefore, findings suggesting lower methane flux in the far past do not necessarily
193 imply lower preindustrial and present-day NMHC emissions. However, if the finding⁵¹ was
194 applied via downscaling to estimate ethane emissions, it then fits poorly with the findings of our
195 preindustrial simulations (Figure 2a), unless we change the geographical distribution towards a
196 larger fraction of emissions in the Northern Hemisphere. As noted (see Methods) the uncertainty
197 in geographical distribution is large.

198 The relative uncertainties in global total emissions in the new fugitive fossil fuel datasets
199 (applied in the ALT1, ALT2 inventories) are about half of those used in CEDS CMIP6 (baseline
200 inventory) (see Supplementary). Another improvement is substantially reduced uncertainties in
201 geographical emission distributions (see Methods). Based on the comparisons in the previous
202 paragraphs the correct global total anthropogenic ethane emissions seem to be close to the levels
203 in the ALT1 and ALT2 simulations (rectangle, triangle Figure S11). These levels are greater than
204 the upper cap of the baseline uncertainty bar in Figure S11, i.e. the baseline inventory likely
205 underestimates emissions. The total emissions in the baseline inventory are close to the mean and
206 median of those in eight other standard community emissions datasets suggesting that applying

207 these will also result in too low modeled ethane concentrations. For the ALT1 and ALT2
208 inventories, only total emissions near the lower end of the uncertainty range (Figure S11)
209 reproduce the observed levels. Various model uncertainties widen the possible emission range
210 but major ones associated to the OH sink have relatively small impacts on modeled ethane
211 concentrations. Model uncertainties do therefore not change our conclusions regarding under-
212 and overestimation in the different emission inventories. Using the alternative emission datasets
213 (ALT1 or ALT2) instead of the standard community emission inventories greatly improves the
214 comparison with observations. Due to sparse observation coverage in some world regions, and
215 the uncertainty ranges of the emissions and atmospheric chemical loss of ethane and propane, we
216 do not provide an overall performance ranking between the emission datasets ALT1 and ALT2.

217

218 **Impacts on other atmospheric constituents**

219 The higher ethane and propane in the ALT1 and ALT2 simulations compared to the baseline
220 simulation impact the greenhouse gas methane and major surface pollutants. The impacts on
221 tropospheric methane are moderate, leading to 0.5 % (ALT1) and 0.7 % (ALT2) higher methane
222 lifetimes due to lower tropospheric hydroxyl. Modeled baseline ozone mixing ratios are
223 compared to surface measurements in Figure S12 for the period June-August when ozone
224 photochemistry is most active in the Northern Hemisphere. The model reproduces the gradients
225 between regions with high photochemical production and cleaner background areas. At many
226 stations the model is at, or within, a few ppb (or percent) of the measurements. In regions with
227 high levels of surface ozone in the baseline, particularly the Middle East and eastern Asia, ozone
228 is 5-13 % (3-11 ppb) higher in spring/summer (Figure S13) in ALT1 and ALT2. In these regions,
229 the ozone production is more sensitive to the amount of NMHCs since high NO_x is present.
230 Surface ozone differences in other regions are generally small (0-5 % or 0-3 ppb). If fossil
231 emissions of other related NMHCs are underestimated as well (e.g. butane, pentane etc.), the
232 impacts on ozone and other air pollutants will be larger. Impacts on the air pollutants NO₂, PAN
233 and CO are discussed in the Supplementary.

234

235 Methane constitutes the largest share of hydrocarbons emitted from fossil sources, and a recent
236 study⁵² suggests underestimation of fossil (geologic + fossil fuel) methane emissions in previous
237 estimates. Compared to previous inventories the much higher fossil fuel ethane and propane
238 emissions in the new datasets^{9,10,35,36} used in ALT1 and ALT2 in this study are mainly due to
239 higher NMHC to methane emission ratios. The improved agreement with ice-core ethane
240 measurements for the simulation with geologic emissions supports the idea that there is a
241 considerable geologic methane emission source^{52,53}. As for ethane and propane, geologic
242 emissions of methane have been neglected in many model studies. In accordance with ref. 52 we
243 suggest a need for more studies evaluating the reported level of fossil (natural geologic +
244 anthropogenic fossil fuel) methane emissions in current emission inventories. Understanding the
245 contribution from different natural and anthropogenic emission sources is a critical precursor to
246 design efficient measures to reverse ongoing atmospheric ethane, propane, and methane
247 increases^{4,54,55}.

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399 **Acknowledgements**

400 This research is funded by the Research Council of Norway through the MOCA (Methane
401 Emissions from the Arctic Ocean to the Atmosphere: Present and Future Climate Effects) project
402 (Grant no. 225814). Furthermore, the conclusions of the paper is largely supported and
403 strengthened by the use of globally distributed observational data and we acknowledge all data
404 providers and the great efforts of EMEP, ACTRIS, NOAA ESRL/INSTAAR, and The World
405 Data Centre for Greenhouse Gases (WDCGG) under the WMO-GAW program to make long-
406 term measurements public and available. The Horizon 2020 research and innovation program
407 ACTRIS-2 Integrating Activities (grant agreement No. 654109) is acknowledged for the work
408 with quality assurance and quality control of NMHC data in Europe. The VOC observations
409 within the NOAA-INSTAAR GGGRN are supported in part by the U.S. National Oceanic and
410 Atmospheric Administration's Climate Program Office's AC4 Program, and quality control was
411 supported in part by the U.S. National Science Foundation grant 1108391.

412 The GLOGOS dataset was kindly provided by CGG Geoconsulting. CGG also provided us with
413 a derived product from the Global Offshore Seepage Database (GOSD) indicating where
414 offshore seepage occurs.

415

416 **Author contributions**

417 S.B.D., G.M., and Ø.H. designed the study with input from A.S., C.L.M., and I.P. S.B.D
418 performed the simulations with the OsloCTM3 model, analyzed the model results, and
419 performed the comparisons with measurement data. Ø.H. and G.M. provided assistance with the
420 analysis and comparison studies. I.P. performed the simulations with the Flexpart model and I.P.
421 and A.S. analyzed the output. S.SC. and L.H.I. provided the new emission datasets for fugitive
422 fossil fuel emissions. S.B.D. developed gridded inventories for geologic emissions. C.L.M, D.H.,
423 S.R., S.SA., N.S., K.A.R., L.J.C., A.C.L., S.P., and M.W. provided the observational data for
424 ethane and propane. S.B.D. led the writing of the manuscript in close collaboration with G.M.
425 and Ø.H. All authors contributed to the writing and review of the manuscript.

426 **Competing financial interests**

427 The authors declare no competing financial interests.

428

429 **Figure captions**

430 **Figure 1: Global total sectoral ethane emissions in this study and other studies. a)** Global
431 total ethane emissions in preindustrial simulations in this study (black symbols) compared to
432 emissions in other studies (blue and yellow symbols and bars). *See Methods section for details.
433 ^δBox model study³⁹. [†]Bottom up inventory⁴⁰. **b)** Global total ethane emissions in the year 2011
434 baseline and alternative (ALT1, ALT2) simulations in this study (black symbols) compared to
435 emissions in other studies. The inventories and estimates cover the year 2000 and onwards. The
436 closest year to our simulation year 2011 is chosen for inventories not covering that year. Bottom
437 up inventories (yellow symbols and bars)[#]: Fossil fuel, biofuel, agriculture, waste: CEDS
438 CMIP6⁴⁷ (used in baseline in this study), HTAPv2, Edgar 3.2 FT, RETRO, POET, CMIP5
439 (average of MACCITY, ACCMIP, RCP2.6, RCP4.5, RCP6, RCP8.5) (as reported and
440 referenced in ECCAD database: <http://eccad.aeris-data.fr/>), ARCTAS (as reported by ref. 7),
441 EDGAR4.3.2 (as reported by ref. 11), and new inventory in ref. 56. Biomass burning: GFEDv4
442 (as used in baseline in this study), GICC, ACCMIP, POET, GFASv1.2, MACCITY, RETRO,
443 RCP2.6, RCP4.5, RCP8.5 (as reported and referenced in ECCAD database: [http://eccad.aeris-](http://eccad.aeris-data.fr/)
444 [data.fr/](http://eccad.aeris-data.fr/)), and FINN (as reported by ref. 3,4). Oceans: RETRO (used in baseline in this study).
445 Vegetation: MEGAN-MACC (used in baseline this study), and MEGANv2 (as reported and
446 referenced in ECCAD database: <http://eccad.aeris-data.fr/>). Natural geologic: As reported by ref.
447 40 (median estimate used in baseline in this study). Top-down estimates from box models
448 (dotted blue bars)^φ: Fossil fuel, biofuel, agriculture, waste: ref. 31,33,39. Biomass burning: ref.
449 39. Geologic: ref. 39. Optimized emissions in 3D model studies (brown symbols)*: Ref. 3,4.

450 **Figure 2: Observed and modeled annual mean ethane mixing ratios and inter-polar ratios. a)**
451 Observed³⁹ and modeled (this study) preindustrial inter-polar ratio and mixing ratios at Summit,
452 Greenland and at the West Antarctic Ice Sheet (WAIS) site. Observation error bars are the reported ± 2
453 standard errors in ref. 39. **b)** Observed and modeled inter-polar ratio and mixing ratios in 2011. For the
454 Antarctic, the closest station (South Pole) with data to the WAIS measurement site (no data for 2011) was
455 used. See Table 1 for more information about the simulations.

456

457 **Figure 3: Comparison of year 2011 modeled and observed ethane and propane at surface**

458 **sites. a)** Comparison of modeled and observed year 2011 ethane (upper row) and propane (lower

459 row) at the Zeppelin station. A selection of comparisons for other sites is shown in the

460 Supplementary. **b)** Comparison of modeled (background colors) and observed surface (color-

461 filled circles) of ethane (ppt) for the year 2011. Model data for the lowest model layer were used.

462 Stations with less than 6 samples within the 3 months averaging period were excluded from the

463 comparison. Mountain stations typically sampling free tropospheric air and situated in areas

464 where the model resolution does not resolve the terrain were also excluded. Details on the

465 applied observation datasets are provided in the Methods section. Maps for the ALT2 simulation

466 are shown in the Supplementary. **c)** Same for propane.

467

468 **Figure 4: Footprints at Zeppelin.** Yearly mean Flexpart footprints (see Methods for details on

469 approach) for ethane at Zeppelin. Upper left: Baseline simulation. Episodes with Largest

470 Underestimation (LU). Upper right panel: ALT1 simulation. Episodes with LU. Lower left:

471 ALT1 simulation. Episodes with Largest Overestimation (LO). Lower right: ALT2 simulation.

472 Episodes with LO. The unit ns/kg is proportional to the residence time in a given volume of air.

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483 **Methods**

484 **Models:**

485 *OsloCTM3:*

486 We use the OsloCTM3 model⁴⁶ to simulate the preindustrial (year 1750) and current (year 2011)
487 distributions of atmospheric ethane and propane. The model is run with approximately 1.1°x1.1°
488 (T159) horizontal resolution. To spin up the model 15 months simulations were made with
489 coarse resolution (2.2°x2.2°) followed by 4 months with 1.1°x1.1° (T159) resolution. After the
490 spin up a set of simulations (Table 1) were made. A coupled tropospheric and stratospheric 60
491 layer (surface-0.1 hPa) version is used with 100 chemical active species affecting atmospheric
492 oxidation capacity. OsloCTM3 was described and evaluated by ref. 46 and was used in several
493 studies related to atmospheric oxidation capacity⁵⁷.

494 The OsloCTM3 simulations are driven with 3-hourly year 2011 meteorological forecast data
495 from the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast
496 System (IFS) model (see ref. 46 for details). These data are 36-hours forecasts produced with 12
497 hours of spin-up starting from an ERA-Interim analysis at noon on the previous day.

498 *Flexpart:*

499 To investigate the origin of air masses observed at the Zeppelin station, we use version 9.2 of the
500 LPDM FLEXPART (FLEXible PARTicle dispersion model)⁴⁸. The model is driven with 3-
501 hourly operational meteorological analyses from the European Centre for Medium-Range
502 Weather Forecasts (ECMWF) with 91 vertical levels and a horizontal resolution of 1°x1°.
503 Computational particles released from the location of the Zeppelin station are tracked 20 days
504 back in time in FLEXPART's "retroplume" mode. The model output consists of an emission
505 sensitivity, the surface footprint of which is used here to identify source regions related to
506 OsloCTM3 under- and overestimation of observed ethane concentrations.

507

508 **Emissions and model simulations**

509 *Baseline emission inventories all constituents*

510 For anthropogenic SO_x, NH₃, CO, NO_x, and NMHCs emissions, we use the Community
511 Emissions Data System (CEDs) Project emission inventory⁴⁷ for the years 1750 and 2011.
512 CEDs is the state of the art dataset currently used in the Coupled Model Inter-comparison
513 Project Phase 6 (CMIP6). For biomass burning, we use GFEDv4⁵⁸ year 2011 emissions and the
514 historical biomass burning dataset for CMIP6 for 1750⁵⁹. Sulfur emissions from other sources are
515 described in ref. 60. Non-methane volatile organic compounds (NMVOCs) emissions from
516 vegetation and oceans are neglected (set to zero) in some studies but not in this study. Biogenic
517 emissions of CO and NMVOCs from vegetation are set to year 2010 (last year covered by
518 dataset) emissions from MEGAN-MACC⁶¹ both for the preindustrial and year 2011 simulations.
519 For NO_x from soil and CO and NMHCs from the oceans we use the year 2000 emissions in the
520 RETRO inventory⁶². Lightning NO_x emissions are described in ref. 46. For natural NH₃ sources
521 we use emissions from GEIA⁶³ for 1990. Methane emissions are described in ref. 57.

522 *Geologic emissions of ethane and propane*

523 For ethane and propane we include geologic emissions suggested by ref. 40 in the baseline
524 emission inventories. For both 1750 and 2011 the global total emissions are set to the medians (3
525 Tg/yr for ethane, 1.7 Tg/yr for propane) of the ranges (2-4 Tg/yr ethane, 1-2.4 Tg/yr propane)
526 estimated by ref. 40. Their study splits the emissions into six main geologic sources: Mud
527 volcanoes, gas seeps, microseepage, submarine seeps, geothermal manifestations and volcanoes.
528 The geographical distribution of geologic emissions has not been gridded to files suitable for
529 atmospheric modeling studies. Here we use several datasets to develop gridded inventories. The
530 emissions from gas seeps (macroseepage) and mud volcanoes are distributed in accordance with
531 the GLOGOS dataset which lists more than 2000 terrestrial (onshore) seeps from 87 countries.
532 In addition to site locations, GLOGOS provides measured or estimated (visually) fluxes of
533 methane for a few sites and methane, ethane and propane concentrations in the gas for some
534 more sites. However, the majority of sites lack such information and we therefore distribute the
535 emissions evenly on the sites to obtain the global total macroseepage emissions estimates from
536 ref. 40. For submarine seeps we used a derived product (see acknowledgement) from the Global
537 Offshore Seepage Database (GOSD) indicating where offshore seepage occurs. We scale the
538 density map from this dataset to obtain the global total emissions from marine seepage in ref. 40.
539 We assume zero emissions from marine seepage in grid-boxes fully covered by sea-ice. For

540 emissions from microseepage we use the CGG Robertson Tellus Sedimentary Basins of the
541 World Map to distribute the emissions from ref. 40. Microseepage, which is diffuse exhalation
542 from soil in petroleum basins, is the largest geologic source but also the most uncertain one^{40,64}
543 in terms of magnitude and geographical distribution. We spread the emissions evenly over the
544 world's petroleum basins, which represents the potential area for such diffusion and we likely to
545 some degree overestimate the geographical extent. We assume that permafrost or thick ice- and
546 snow-layers hinder diffusion. The northward and southward extent of emissions is therefore
547 limited to 66° N and 60° S to account for this in a simplified way. The emissions from volcanoes
548 and geothermal sources are gridded using the geographical distribution for SO₂ emissions⁶⁰ for
549 such activity. Geologic seepage (macroseepage, some microseepage, and marine seepage) occur
550 at many of the places where current oil, gas, and coal extraction take place.

551

552 *Baseline and alternative year 1750 preindustrial ethane emissions and simulations*

553 The preindustrial baseline simulation (Table 1) includes geologic emissions of the magnitude
554 suggested by Etiope et al.⁴⁰ and within the range found by Nicewonger et al.³⁹ (Figure 1a). In
555 Nicewonger et al. an observed inter-polar ethane asymmetry requires a certain combination of
556 emissions from biomass burning and natural geologic sources (Figure 1a). Preindustrial biomass
557 burning emissions are particularly uncertain, and the magnitude in the inventory (CEDS CMIP6
558 year 1750) in our baseline simulation is high compared to the range obtained by ref. 39. We
559 therefore perform an additional simulation using a biomass burning inventory (CMIP5 1850⁶⁵)
560 with lower emissions (Table 1, Figure 1a) and different geographical distribution. Transport to
561 high southern latitudes has inter-annual variability. To check the sensitivity on our results we do
562 a simulation with meteorological input data for a different year (Table 1).

563

564 *Baseline year 2011 ethane and propane emissions and simulation*

565 Figure 1b shows global total sectoral ethane emissions used in the year 2011 baseline simulation
566 in this study compared to emissions used in other inventories. Based on the preindustrial
567 simulations we include geologic emissions in our baseline simulation for current conditions (year
568 2011). Except for the box model optimized inventory for 2000-2010 in Nicewonger et al.,

569 geologic emissions were not included in any other model studies. Our applied emissions for
570 biomass burning (GFEDv4 year 2011) are in the mid-range compared to other inventories. The
571 other anthropogenic emissions (sum of fossil fuel, biofuel, agriculture, and waste in Figure 1b) in
572 the CEDS CMIP6 data⁴⁷ applied in this study are quite close to the median of the estimates in
573 other inventories. Due to likely trends in anthropogenic emissions over the last decades^{3,4,31,33},
574 different basis years partly explain the large range of the emission estimates in various
575 inventories presented in Figure 1b. The other reason is the large uncertainties in existing
576 inventories due to incomplete approaches and datasets (discussed in main text and section
577 below). Summing up all sectors in Figure 1b our total emissions are in the upper range of other
578 studies. This is mainly due to the inclusion of geologic emissions.

579

580 *Alternative year 2011 ethane and propane emissions and simulations*

581 We also perform simulations with alternative ethane and propane emissions from the energy
582 sector (Table 1). In these we replace emissions from the energy sector in the CEDS CMIP6
583 inventory with two new datasets^{9,10,35,36} for upstream (fuel production, gathering, and processing)
584 and downstream (transmission and distribution) emissions from oil, gas and coal systems. These
585 studies take much better into account that the emission factors from venting and flaring of
586 associated gas released during extraction vary considerably across different oil, coal and gas
587 fields in the world. The studies used novel approaches to quantify and attribute methane and
588 NMHC emissions, combining field measurements and country specific information from
589 published sources with observed flaring of associated gas from satellite images, to arrive at
590 country-specific emissions from flows of associated gas. Two simulations are performed with
591 these datasets. In what we refer to as the ALT1 simulation emissions from oil and gas from ref.
592 10 are combined with coal emissions from ref. 9 (updated with data from ref. 36) to obtain a
593 complete inventory for the energy sector. In the simulation entitled ALT2 we use oil, gas, and
594 coal emissions from ref. 9,35 updated with data from ref. 36. The dataset does not include
595 propane emissions and we use the global mean propane to ethane emission ratio from the ref. 10
596 datasets to obtain propane emissions for the ALT2 simulation. In ALT2 we use the low estimate
597 for natural gas from ref. 9 since it was shown to be the most likely³⁵. Table 2 provides an
598 overview of the fugitive fossil fuel emissions in the ALT1 and ALT2 inventories. Due to

599 substantial geographical overlap it is likely that some emissions from geologic seepage are
600 included³⁵ in fugitive fossil fuel inventories. To avoid double-counting, we subtract ethane and
601 propane emissions from the oil, gas, and coal grids. In the absence of a well-established gridded
602 emission inventory of geologic seeps, we subtract geologic seepage ethane and propane
603 emissions from oil, gas, and coal grids in equal parts, i.e., one third each. By reducing with
604 amounts corresponding to all emissions from geologic seepage, the resulting inventories (ALT1
605 and ALT2) could be regarded as lower estimates of emissions from current oil, gas, and coal
606 activity relative to ref. 10 and ref. 9.

607

608 *Uncertainties in baseline and alternative anthropogenic emission inventories*

609 The uncertainty calculations for the baseline inventory and alternative (ALT1, ALT2) inventories
610 are discussed in the Supplementary in relation to Figure S11.

611

612 *Sensitivity simulation on atmospheric sink*

613 Oxidation by hydroxyl (OH) in the troposphere is the main sink for ethane and propane^{1,2,41}. The
614 uncertainty for the reaction rates are rather small, 15-20 % at 298 K^{2,66}. For hydroxyl
615 concentrations, the uncertainty is larger. Our global averaged tropospheric OH (1.35×10^6
616 molecules/cm³) in our 2011 baseline simulation is on the high side compared to other model
617 studies⁴⁵. The same holds for the hemispheric OH ratio (1.55) compared to model and
618 observational based estimates^{42,44}. The modeled global average methane lifetime, which is highly
619 dependent on the modeled OH concentration, is also low compared to observational based
620 estimates^{67,68}. We therefore did a sensitivity study scaling down the global mean OH
621 concentration to 1×10^6 molecules/cm³. The scaling was done separately for the hemispheres to
622 also get a hemispheric ratio of 1. This is in the lower range of model and observation based
623 estimates both for the OH concentration and hemispheric ratio and was done to see to what
624 degree lower OH concentrations in the Northern Hemisphere would improve the comparison
625 with observations. The OH concentration was scaled down only in the chemical reactions with
626 ethane and propane and not in reactions with other atmospheric constituents.

627 **Measurement data**

628 More and more high quality measurements of NMHC have become available through
629 coordination by the World Meteorological Organization (WMO) Global Atmospheric Watch
630 (GAW) program. Participating networks include ACTRIS (Aerosol, Clouds, and Trace gases
631 Research Infrastructure, the European Research Infrastructure for the observation of Aerosol,
632 Clouds, and Trace gases (<http://www.actris.eu>)), EMEP (The European Monitoring and
633 Evaluation Program) and NOAA ESRL/INSTAAR (National Oceanic & Atmospheric
634 Administration - Earth System Research Laboratory/Institute for Arctic and Alpine Research).
635 To reveal strengths and discrepancies in model performance and evaluate emission inventories
636 we compare the model results to surface ethane and propane observations for the year 2011. We
637 use data from surface sites reported to the World Data Center for Greenhouse Gases (WDCGG)
638 (<http://ds.data.jma.go.jp/gmd/wdcgg/>) following the NOAA/INSTAAR and WMO-GAW
639 measurement protocols, and from EMEP complying with ACTRIS recommendations. EMEP and
640 ACTRIS data were downloaded from EBAS (<http://ebas.nilu.no>), and are also accessible through
641 the ACTRIS data portal (<http://actris.nilu.no>). These data follow the ACTRIS quality assurance
642 procedures and protocols. Generally, the ACTRIS data have higher time resolution (up to 2
643 hour). Intercomparison exercises⁶⁹⁻⁷¹ have shown consistency of NOAA/INSTAAR and
644 EMEP/ACTRIS measurement sites within the data quality objective of +/-10% of the WMO
645 Global Atmospheric Watch program^{72,73}.

646 A subset of data from 96 sites from a total of 132 sites (66 for both ethane and propane) are
647 shown in the comparisons for 2011. The observations at the given location, altitude and time
648 were compared to output for the closest model grid box, level, and time. Figure 3 shows all sites
649 and Figure S4 shows the locations of the subset of sites selected for detailed comparison to
650 model results. Criteria for selection were data quality assurance, access to continuous time series
651 with few gaps, coverage of different regions, and site characteristics (e.g. elevation, topography,
652 and influence of pollution episodes) likely captured by the resolution of a global model.

653 A comparison between modelled and observed ozone is performed in the Supplementary and the
654 applied ozone measurement data are presented there.

655

656

657 **Table 1:** Overview of the simulations performed with OsloCTM3.

Year	Simulation	Simulation name	Inventories anthropogenic and natural emissions*	Geologic emissions	Oxidation chemistry	Meteorological year
1750	Preindustrial	1750 Baseline	Baseline	Yes	Interactive	2011
	Preindustrial sensitivity	1750 NOGEO	Baseline	No	Interactive	2011
		1750 MET	Baseline	Yes	Interactive	2014
		1750 BBURN	Baseline, but CMIP5 biomass burning	Yes	Interactive	2011
2011	Baseline	2011 Baseline	Baseline	Yes	Interactive	2011
	Baseline sensitivity	OH	Baseline	Yes	Interactive, OH field scaled down in reactions with ethane and propane**	2011
	Alternative	ALT1	ALT1	Yes	Interactive	2011
		ALT2	ALT2	Yes	Interactive	2011

658 *The baseline, ALT1 and ALT2 emission inventories are described in the above section.

659 ** The scaling of OH is described in the above section.

660

661 **Table 2:** Overview of the new fugitive fossil fuel emission inventories used in the alternative
662 year 2011 simulations.

Inventory	Natural gas	Oil	Coal
ALT1 ethane	ref. 10	ref. 10	ref. 9 updated with data from ref. 36
ALT1 propane	Same as above	Same as above	Same as above
ALT2 ethane	ref. 9,35 updated with data from ref. 36	ref. 9 updated with data from ref. 36	ref. 9 updated with data from ref. 36
ALT2 propane	Same as above using propane to ethane ratio from ref. 10	Same as above using propane to ethane ratio from ref. 10	Same as above using propane to ethane ratio from ref. 10

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672 **Code availability.** We have opted not to make the computer codes associated with this paper
673 available, because replication of our results does not require access to the computer codes.

674 **Data availability.** The ethane and propane surface measurement data used in this study are
675 freely available. We use data reported to the World Data Center for Greenhouse Gases
676 (WDCGG) (<http://ds.data.jma.go.jp/gmd/wdcgg/>). EMEP and ACTRIS data are available from
677 EBAS (<http://ebas.nilu.no>), and are also accessible through the ACTRIS data portal
678 (<http://actris.nilu.no>). The sites used for detailed comparison with model results are listed in
679 Table S1. The sites can easily be found by name or map search in the databases. The new
680 emission datasets for fugitive fossil fuel emissions are available upon request to Lena Höglund-
681 Isaksson (hoglund@iiasa.ac.at) and Stefan Schwietzke (stefan.schwietzke@noaa.gov). The
682 gridding used to develop geologic ethane and propane emissions suitable for atmospheric
683 modeling studies was based on commercial datasets owned by CGG geoconsulting. CGG should
684 be contacted and consulted for the task of gridding geologic emissions.

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