

**Supplementary Materials for: Bayesian assessment of chlorofluorocarbon (CFC), hydrochlorofluorocarbon (HCFC) and halon banks suggest large reservoirs still present in old equipment.**

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**Summary of Prior distributions**

Below, we outline prior assumptions for each of the input parameters in the simulation model, parameters in the likelihood function, and the timeframe for the simulation model and observational data. Mean estimates of the priors for each of the input parameters are derived from AFEAS estimates, or previously published data, as indicated below. Large uncertainties are specified for each of the prior distributions. If the prior distributions resulted in simulated mole fractions that did not encompass observed mole-fractions then prior uncertainties were increased until observed mole fractions were within uncertainty of the prior mole fraction distributions.

**HCFC-22 model specifications**

Equipment Categories	Short bank	Medium bank	Long bank	Feedstock
<b>Release Functions</b>				
Year 1 emissions:  Prior Distribution	83% of production  1 – Year 2 distribution	As a fraction of production: 1944-1977: 20% 1977-1984: 10% 1984-1993: 7% 1993-2019: 6%  Beta Distribution with parameters 6, 6 centered on above values with lower bounds = 0, and upper bound twice the value.	NA	Release of 2% of feedstock production  Lognormal distribution such that: $\mu = 2\%$ $\sigma = 1\%$
Subsequent Year emissions:  Prior Distribution	Year 2: 17% of production.  Beta Distribution with parameters 4, 4, centered on 17% such that: $\mu = 17\%$ $\sigma = 6\%$	Release of 10% of bank a year  Lognormal distribution such that: $\mu = 10\%$ $\sigma = 5\%$	Release of 2% of bank a year  Lognormal distribution such that: $\mu = 2\%$ $\sigma = 1\%$	NA

<p><b>Production</b></p> <p>Developed using AFEAS data (<a href="https://agage.mit.edu/sites/default/files/documents/em-hcfc-22.pdf">https://agage.mit.edu/sites/default/files/documents/em-hcfc-22.pdf</a>) to partition production into short, medium and long banks. Production is then scaled to match UNEP total production estimates for A5 and non-A5 countries. Following 2003, AFEAS data is no longer available and we assume total production is discretized into the AFEAS categories at the same proportion as is reported in 2003. Amounts of HCFC-22 used for feedstock production are available annually (UNEP/TEAP, 2021). A prior mean leakage rate of 2% was assumed during production, which reflects a medium value between different facilities (MCTOC, 2019). For each of the uses, production follows a log normal distribution built on reported production, <math>Prod_{0,t}</math>, such that yearly production values are;</p> $Prod_t \sim Prod_{0,t} \times (0.2 * Log(0, 0.5) + 0.90)$ <p>Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1.</p>
<p><b>Details of Simulation Model</b></p> <p>The simulation model is run between 1944 – 2019. Each equipment type is simulated separately (short, medium, long), and total banks the sum of the three. Emissions are the sum of the three equipment type emissions along with feedstock emissions which have a leakage rate prior distribution specified above.</p>
<p><b>Likelihood Function</b></p> <p>Posteriors are conditioned on observed mole fractions between 2004– 2019. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a beta distribution with lower bound at 3% and upper bound at 6% of observed mole fractions. A correlation of R = 0.98 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.</p>

### HCFC-141b model specifications

Equipment Categories	Short bank	Medium bank	Long bank
<b>Release Functions</b>			
Year 1 emissions:	83% of production	30% of production	10% of production
Prior Distribution	1 – Year 2 distribution	Beta Distribution with parameters 4, 4 centered on 0.3 with lower bounds = 0.15, and upper bound = 0.45	Beta Distribution with parameters 4, 4 centered on 0.1 with lower bounds = 0, and upper bound = 0.2
Subsequent Year emissions:	Year 2: 17% of production.	4.5 year lifetime or ~ 20%/year loss rate.	Release of 4.5% of bank a year
Prior Distribution	Beta Distribution with parameters 4, 4, centered on 17% such that: $\mu = 17\%$ $\sigma = 6\%$	Beta distribution with parameters 4,4 centered on 0.2, with lower bound 0 and upper bound 0.4	Lognormal distribution such that: $\mu = 4.5\%$ $\sigma = 0.9\%$
<b>Production</b>			
Developed using AFEAS data ( <a href="https://agage.mit.edu/sites/default/files/documents/em-hcfc-141b.pdf">https://agage.mit.edu/sites/default/files/documents/em-hcfc-141b.pdf</a> ) to partition production into short, medium and long banks. For each of the uses, production follows a log normal distribution built on reported production, $Prod_{0,t}$ , such that yearly production values are;			

$$Prod_t \sim Prod_{0,t} \times (0.2 * Log(0, 0.5) + 0.90)$$

Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1. No feedstock production is included in the analysis.

**Details on Simulation model**

The simulation model is run between 1944 – 2019. Each equipment type is simulated separately (short, medium, long), and total banks and emissions are the sum of the three.

**Likelihood Function**

Posteriors are conditioned on observed mole fractions between 2004 – 2019. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a beta distribution with lower bound at 3.5% and upper bound at 6% of observed mole fractions. A correlation of R = 0.95 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

**HCFC-142b model specifications**

Equipment Categories	Short bank	Medium bank	Long bank	
<b>Release Functions</b>				
Year 1 emissions:	83% of production	30% of production	32.5% of production	2% of feedstock production.
Prior Distribution	1 – Year 2 distribution	Beta Distribution with parameters 4, 4 centered on 0.3 with lower bounds = 0.15, and upper bound = 0.45	Beta Distribution with parameters 4, 4 centered on 0.325 with lower bounds = 0.15, and upper bound = 0.5	Lognormal distribution such that: $\mu = 2\%$ $\sigma = 1\%$
Subsequent Year emissions:	Year 2: 17% of production.	4.5 year lifetime or ~ 20%/year loss rate.	Release of 4.5% of bank a year	NA
Prior Distribution	Beta Distribution with parameters 4, 4, centered on 17% such that: $\mu = 17\%$ $\sigma = 6\%$	Beta distribution with parameters 4,4 centered on 0.2, with lower bound 0 and upper bound 0.4	Lognormal distribution such that: $\mu = 3\%$ $\sigma = 0.6\%$	

**Production**

Developed using AFEAS data (<https://agage.mit.edu/sites/default/files/documents/em-hcfc-142b.pdf>) to partition production into short, medium and long banks. Amounts of HCFC-142b used for feedstock production are available annually (UNEP/TEAP, 2021). A prior mean leakage rate of 2% was assumed during production, which reflects a medium value between different facilities (MCTOC, 2019). For each of the uses, production follows a log normal distribution built on reported production,  $Prod_{0,t}$ , such that yearly production values are;

$$Prod_t \sim Prod_{0,t} \times (0.3 * Log(0, 0.5) + 0.95)$$

Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1.

<b>Details on Simulation model</b>
The simulation model is run between 1944 – 2019. Each equipment type is simulated separately (short, medium, long), and total banks and emissions are the sum of the three.
<b>Likelihood Function</b>
Posteriors are conditioned on observed mole fractions between 2004 – 2019. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard that is inferred from a beta distribution with lower bound at 2.8% and upper bound at 5% of observed mole fractions. A correlation of R = 0.96 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

### Halon-1211 model specifications

Equipment Categories	Single category
<b>Release Functions</b>	
Year 1 emissions:	Prior to 1990: 20% of production 1990 - present: 5% of production
Prior Distribution	Beta Distribution with parameters 2, 2. Prior to 1990: Centered on 0.2 with lower bounds = 0, and upper bound = 0.4 1990 - present: Centered on 0.05 with lower bounds = 0, and upper bound = 0.1 *Note that these priors reflect large unknowns in direct emissions during production as this hasn't been well documented to our knowledge. This high upper bound comes from McCulloch (1992) that suggests that People's Republic of China lost 30-40% of annual production each year to direct emissions.
Subsequent Year emissions:	Prior to 1990: 6% of production 1990 - present: 4% of production
Prior Distribution	Beta distribution with parameters 2,2 Prior to 1990: Centered on 0.06 with lower bounds = 0, and upper bound = 0.12 1990 - present: Centered on 0.04 with lower bounds = 0, and upper bound = 0.08 *These priors are developed from the HTOC report <a href="https://ozone.unep.org/sites/default/files/assessment_panels/HTOC_assessment_2018.pdf">https://ozone.unep.org/sites/default/files/assessment_panels/HTOC_assessment_2018.pdf</a> as well as values from Chapter 9 from SROC.
<b>Production</b>	
Developed using McCulloch (1992) up to 1990, following 1990, UNEP data is used. McCulloch reports varying levels of uncertainty for different time periods, these are used to develop production priors for three different time periods that are independent from one another: Pre-1980, production follows a log normal distribution built on reported production, $Prod_{0,t}$ , with the widest uncertainties such that yearly production values are;	
$Prod_t \sim Prod_{0,t} \times (0.5 * Log(0, 0.5) + 0.80)$	
1981-2019, uncertainties are smaller:	
$Prod_t \sim Prod_{0,t} \times (0.3 * Log(0, 0.5) + 0.95)$	
Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1. The correlation term is included for three distinct time periods, (pre-1980, 1981 – 1990, 1991 – 2019), to reflect that we expect there to be common biases in reporting within those time periods, but that the correlation does not necessarily apply across time periods when reporting practices change.	

<b>Details on Simulation model</b>
The simulation model is run between 1963 – 2019. Each equipment type is simulated separately (short, medium, long), and total banks and emissions are the sum of the three.
<b>Likelihood Function</b>
Posteriors are conditioned on observed mole fractions between 2004 – 2019. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution with lower bound equal to 4% and upper bound equal to 6% of observed mole fractions. A correlation of R = 0.7 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

### Halon-1301 model specifications

Equipment Categories	Single category
<b>Release Functions</b>	
Year 1 emissions:	Prior to 1990: 20% of production 1990 - present: 5% of production
Prior Distribution	Beta Distribution with parameters 2, 2: Prior to 1990: Centered on 0.2 with lower bounds = 0, and upper bound = 0.4 1990 - present: Centered on 0.05 with lower bounds = 0, and upper bound = 0.1 *As in Halon-1211, these priors reflect large unknowns in direct emissions during production as this hasn't been well documented to our knowledge. This high upper bound comes from McCulloch (1992) that suggests that People's Republic of China lost 30-40% of annual production each year to direct emissions.
Subsequent Year emissions:	Prior to 1990: 5% of production 1990 - present: 4% of production
Prior Distribution	Beta distribution with parameters 2,2 Prior to 1990: Centered on 0.06 with lower bounds = 0, and upper bound = 0.12 1990 - present: Centered on 0.04 with lower bounds = 0, and upper bound = 0.08 *These priors are developed from the HTOC report <a href="https://ozone.unep.org/sites/default/files/assessment_panels/HTOC_assessment_2018.pdf">https://ozone.unep.org/sites/default/files/assessment_panels/HTOC_assessment_2018.pdf</a> as well as values from Chapter 9 from SROC.
<b>Production</b>	
Developed using McCulloch (1992) up to 1990, following 1990, UNEP data is used. McCulloch reports varying levels of uncertainty for different time periods, these are used to develop production priors for three different time periods that are independent from one another: Pre-1972, production follows a log normal distribution built on reported production, $Prod_{0,t}$ , with the widest uncertainties such that yearly production values are;	
$Prod_t \sim Prod_{0,t} \times (0.5 * Log(0, 0.5) + 0.80)$	
1973-2019, uncertainties are smaller:	
$Prod_t \sim Prod_{0,t} \times (0.3 * Log(0, 0.5) + 0.95)$	
Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1. The correlation term is included for three distinct time periods, (pre-1980, 1981 – 1990, 1991 – 2019), to reflect that we expect there to be common biases in reporting within those time periods, but that the correlation does not necessarily apply across time periods when reporting practices change.	

<b>Details on Simulation model</b>
The simulation model is run between 1963 – 2019. Each equipment type is simulated separately (short, medium, long), and total banks and emissions are the sum of the three.
<b>Likelihood Function</b>
Posteriors are conditioned on observed mole fractions between 2004 – 2019. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution with lower bound equal to 3% and upper bound equal to 6% of observed mole fractions. A correlation of $R = 0.7$ is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

### CFC-11

Equipment categories	Non-hermetic Refrigeration	Blowing Agents Closed Cell Foam	Open Cell Foam, Aerosols and Others
<b>Release Fractions</b>			
Year 1:	Year 1: 7% release	NA	Year 1: 83% release for Open Cell Foam, 50% release for Aerosols
Prior Distribution	Lognormal with parameters such that $\mu = 7\%$ and $\sigma = 3.5\%$		Beta, parameters 8, 6 which gives: $\mu = 57\%$ , $\sigma = 13\%$
Subsequent years:	Life years: 10 years	3.66% of bank	Year 2: 17% release for open cell foam, 50% release for aerosols
Prior Distribution	Lognormal $\mu = 10$ years, $\sigma = 2$ year	Lognormal with parameters such that $\mu = 3.66\%$ and $\sigma = 1.83\%$	Modeled jointly with Year 1 distribution. (1 – Year 1 release)
<b>Production</b>			
Developed using AFEAS data up until 1989 ( <a href="https://unfccc.int/files/methods/other_methodological_issues/interactions_with_ozone_layer/application/pdf/cfc1100.pdf">https://unfccc.int/files/methods/other_methodological_issues/interactions_with_ozone_layer/application/pdf/cfc1100.pdf</a> ) and augmented by UNEP data following 1989. AFEAS data is used to partition production into the various equipment types. For each of the uses, production follows a log normal distribution built on reported production, $Prod_{0,t}$ , such that yearly production values are;			
$Prod_t \sim Prod_{0,t} \times (0.2 * Log(0, 0.5) + 0.95)$			
Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1. Following (Lickley et al., 2020), we address the unexpected emissions, as identified by (Montzka et al., 2018), by adding an unreported production term that has a linearly increasing upper bound starting at 0 in 2000 and increasing to 61,000 tonnes in 2012, after which point the upper bound is held constant through the remainder of the simulation period. The distribution following 2000 is a uniform distribution between 0 and the upper bound in each year, to reflect the level of uncertainty in production throughout that time period.			
<b>Details on Simulation Model</b>			
The bank simulation model runs from 1955-2018.			
<b>Likelihood Function</b>			
Posteriors are conditioned on observed mole fractions between 1980 – 2018. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution			

with lower bound equal to 2% and upper bound equal to 4% of observed mole fractions. A correlation of  $R = 0.96$  is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

### CFC-12 Model Specifications

Equipment categories	Non-hermetic Refrigeration	Hermetic Refrigeration	Blowing Agents Closed Cell Foam	Open Cell Foam, Aerosols and Others
Year 1:	Year 1: 7% release	Year 1: 2% release	Year 1: 50% release	Year 1: 83% release for Open Cell Foam, 50% release for Aerosols and others
Prior Distribution	Lognormal with parameters s.t. $\mu = 7\%, \sigma = 3.5\%$	Lognormal with parameters s.t. $\mu = 2\%, \sigma = 1\%$	Beta Distribution with parameters 2, 2 which gives: $\mu = 50\%, \sigma = 22\%$	Beta, parameters 2, 2 which gives: $\mu = 50\%, \sigma = 22\%$
Subsequent years:	Life years: 10 years	Life years: 20 years	Year 2: 50% release	Year 2: 50% release
Prior Distribution	Lognormal $\mu = 10 \text{ years}, \sigma = 2 \text{ year}$	Lognormal $\mu = 20 \text{ years}, \sigma = 4 \text{ year}$	Modeled jointly with Year 1. (1 – Year 1 release)	Modeled jointly with Year 1. (1 – Year 1 release)

#### Production

Developed using AFEAS data up until 1989

([https://unfccc.int/files/methods/other\\_methodological\\_issues/interactions\\_with\\_ozone\\_layer/application/pdf/cfc1200.pdf](https://unfccc.int/files/methods/other_methodological_issues/interactions_with_ozone_layer/application/pdf/cfc1200.pdf)) and augmented by UNEP data following 1989. AFEAS data is used to partition production into the various equipment types. For each of the uses, production follows a log normal distribution built on reported production,  $Prod_{0,t}$ , such that yearly production values are;

$$Prod_t \sim Prod_{0,t} \times (0.2 * Log(0, 0.5) + 0.95)$$

Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1.

#### Details on Simulation Model

The bank simulation model runs from 1955-2018.

#### Likelihood Function

Posteriors are conditioned on observed mole fractions between 1980 – 2018. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution with lower bound equal to 1.5% and upper bound equal to 3% of observed mole fractions. A correlation of  $R = 0.98$  is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

### CFC-113 Model Specifications

Equipment categories	Short Bank	Long Bank
Year 1:	Year 1: 50% release	Year 1: 2% release
Prior Distribution	Beta Distribution with parameters 12, 12 which gives: $\mu = 50\%, \sigma = 10\%$	Lognormal with parameters s.t.

		$\mu = 2\%, \sigma = 1\%$
Subsequent years:	Year 2: 50% release	Life years: 20 years
Prior Distribution	Modeled jointly with Year 1. (1 – Year 1 release)	Lognormal $\mu = 20 \text{ years}, \sigma = 4 \text{ year}$
<b>Production</b>		
<p>Developed using AFEAS data up until 1989 (<a href="https://agage.mit.edu/sites/default/files/documents/em-cfc-113.pdf">https://agage.mit.edu/sites/default/files/documents/em-cfc-113.pdf</a>) and augmented by UNEP data following 1989. AFEAS data is used to partition production into the various equipment types. Amounts of CFC-113 used for feedstock production are available annually (UNEP/TEAP, 2021). A prior mean leakage rate of 2% was assumed during production, which reflects a medium value between different facilities (MCTOC, 2019). For short bank and long bank uses, production follows a log normal distribution built on reported production, <math>Prod_{0,t}</math>, such that yearly production values are;</p> $Prod_t \sim Prod_{0,t} \times (0.4 * Log(0, 0.5) + 0.70)$ <p>For CFC-113 feedstock, production follows a log normal distribution;</p> $Prod_t \sim Prod_{0,t} \times (0.3 * Log(0, 0.5) + 0.95)$ <p>Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1.</p>		
<b>Details on Simulation Model</b>		
The bank simulation model runs from 1955-2018.		
<b>Likelihood Function</b>		
<p>Posteriors are conditioned on observed mole fractions between 2004– 2018. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution with lower bound equal to 1% and upper bound equal to 4% of observed mole fractions. A correlation of R = 0.95 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.</p>		

### CFC-114 Model Specifications

Equipment categories	Short Banks	Long Banks
Year 1:	Year 1: 50% release	Year 1: 2% release
Prior Distribution	Beta Distribution with parameters 8, 8 which gives: $\mu = 50\%, \sigma = 12\%$	Lognormal with parameters s.t. $\mu = 2\%, \sigma = 2\%$
Subsequent years:	Year 2: 50% release	Lifetime: 20 years
Prior Distribution	modeled jointly with Year 1. (1 – Year 1 release)	Lognormal bank emissions s.t. $\mu = 4.88\% \text{ release}, \sigma = 2.44\%$
<b>Production</b>		
<p>Developed using AFEAS data up until 1989 (<a href="https://agage.mit.edu/sites/default/files/documents/em-cfc-114.pdf">https://agage.mit.edu/sites/default/files/documents/em-cfc-114.pdf</a>) and augmented by UNEP data following 1989. AFEAS data is used to partition production into the various equipment types.</p>		

For each of the uses, production follows a log normal distribution built on reported production,  $Prod_{0,t}$ , such that yearly production values are;

$$Prod_t \sim Prod_{0,t} \times (0.2 * Log(0, 0.5) + 0.90)$$

Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1.

**Details on Simulation Model**

The bank simulation model runs from 1935-2019.

**Likelihood Function**

Posteriors are conditioned on observed mole fractions between 2004– 2019. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution with lower bound equal to 1% and upper bound equal to 3% of observed mole fractions. A correlation of R = 0.7 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

**CFC-115 Model Specifications**

Equipment categories	Short Bank	Long Bank
Year 1:	Year 1: 50% release	Year 1: 7% release
Prior Distribution	Beta Distribution with parameters 8, 8 which gives: $\mu = 50\%$ , $\sigma = 12\%$	Beta Distribution with parameters 4, 4 centered on 7% which gives: $\mu = 7\%$ , $\sigma = 2.33\%$
Subsequent years:	Year 2: 50% release	Lifetime: 10 years
Prior Distribution	modeled jointly with Year 1. (1 – Year 1 release)	Lognormal bank emissions s.t. $\mu = 9.5\%$ release, $\sigma = 4.75\%$

**Production**

Developed using AFEAS data up until 1989 (<https://agage.mit.edu/sites/default/files/documents/em-cfc-115.pdf>) and augmented by UNEP data following 1989. AFEAS data is used to partition production into the various equipment types. For each of the uses, production follows a log normal distribution built on reported production,  $Prod_{0,t}$ , such that yearly production values are;

$$Prod_t \sim Prod_{0,t} \times (0.2 * Log(0, 0.5) + 0.9)$$

Year-to-year values deviate from reported production with a correlation term drawn from a beta distribution with parameters 2, 2, and lower bound of 0.5 and upper bound of 1.

**Details on Simulation Model**

The bank simulation model runs from 1935-2019.

**Likelihood Function**

Posteriors are conditioned on observed mole fractions between 2004– 2010. The likelihood function is a multivariate normal likelihood function of the difference between yearly observed and modeled mole fractions is used with mean equal to zero and standard deviation that is inferred from a prior following a beta distribution with lower bound equal to 5% and upper bound equal to 8% of observed mole fractions. A correlation of R = 0.3 is included in the multivariate covariance function to represent an assumed correlated error between modeled and observed mole fraction.

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