Multi-species inversion and IAGOS airborne data for a better constraint of continental-scale fluxes

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Abstract. Airborne measurements of CO₂, CO, and CH₄ proposed in the context of IAGOS (In-service Aircraft for a Global Observing System) will provide profiles from take-off and landing of airliners in the vicinity of major metropolitan areas useful for constraining sources and sinks. A proposed improvement of the top-down method to constrain sources and sinks is the use of a multispecies inversion. Different species such as CO₂ and CO have partially overlapping emission patterns for given fuel-combustion-related sectors, and thus share part of the uncertainties related both to the a priori knowledge of emissions and to model–data mismatch error. We use a regional modelling framework consisting of the Lagrangian particle dispersion model STILT (Stochastic Time-Inverted Lagrangian Transport) combined with the high-resolution (10 km × 10 km) EDGARv4.3 (Emission Database for Global Atmospheric Research) emission inventory, differentiated by emission sector and fuel type for CO₂, CO, and CH₄, and combined with the VPRM (Vegetation Photosynthesis and Respiration Model) for biospheric fluxes of CO₂. Applying the modelling framework to synthetic IA-GOS profile observations, we evaluate the benefits of using correlations between different species’ uncertainties on the performance of the atmospheric inversion. The available IA-GOS CO observations are used to validate the modelling framework. Prior uncertainty values are conservatively assumed to be 20 %, for CO₂ and 50 % for CO and CH₄, while those for GEE (gross ecosystem exchange) and respiration are derived from existing literature. Uncertainty reduction for different species is evaluated in a domain encircling 50 % of the profile observations’ surface influence over Europe. We found that our modelling framework reproduces the CO observations with an average correlation of 0.56, but simulates lower mixing ratios by a factor of 2.8, reflecting a low bias in the emission inventory. Mean uncertainty reduction achieved for CO₂ fossil fuel emissions is roughly 38 %; for photosynthesis and respiration flux it is 41 and 44 % respectively. For CO and CH₄ the uncertainty reduction is roughly 63 and 67 % respectively. Considering correlation between different species, posterior uncertainty can be reduced by up to 23 %; such a reduction depends on the assumed error structure of the prior and on the considered time frame. The study suggests a significant uncertainty constraint on regional emissions using multi-species inversions of IAGOS in situ observations.

1 Introduction

As widely recognized at the international level, there is a need for reduction in anthropogenic emissions (IPCC, 2014). This however implies the necessity for reliable climate predictions from atmospheric models in order to allow policymakers to make informed decisions. Unfortunately, current climate predictions are hampered by excessive uncertainties; for example intercomparisons of different models show important differences in their predictions as shown in Friedlingstein et al. (2006). This makes it difficult to assess the better environmental policies to implement. Because most biogenic...
fluxes in Europe are influenced by human activities, with 22 % of Europe’s land dedicated to agriculture (FAO, 2013) and 45 % covered by forests, of which 80 % is managed for wood supply (UNECE, FAO, 2011), understanding and managing these biogenic fluxes must also be a component of any policy to reduce anthropogenic emissions.

A commonly used approach to estimate carbon budgets by teasing apart sources and sinks in a given spatial domain is the atmospheric Bayesian inversion. Atmospheric inversions combine prior knowledge from emission inventories with atmospheric observations acting as a top-down constraint to produce better posterior knowledge. As the main goal of this study is to assess the benefit of inter-species correlations in reducing the uncertainty of the posterior state space, we are particularly interested in the effects of such correlations on the uncertainty reduction, defined as the difference between prior and posterior uncertainty normalized by the prior. The vast majority of published papers on atmospheric inversions investigate the budget of a single species, usually a long-lived greenhouse gas like CO\textsubscript{2} (e.g. Rödenbeck et al., 2003) or CH\textsubscript{4} (e.g. Hein et al., 1997; Bousquet et al., 2006), but the technique can also be applied to active species like CO (Bergamaschi et al., 2000). Note that carbon dioxide is a special case as atmospheric CO\textsubscript{2} inverting ratio result from a combination of strong anthropogenic sources with strong sources and sinks from biospheric processes, calling for a separation of anthropogenic fluxes from biospheric fluxes. One way to achieve such a separation is to measure CO alongside CO\textsubscript{2}, and use CO as a proxy for CO\textsubcript{2} anthropogenic emissions. Several studies have made use the correlations among different species. One of the first examples is the work from Enting et al. (1995) on CO\textsubscript{2} and 13\textsubscript{CO}\textsubscript{2}, while Brioude et al. (2012) attempted to derive a CO\textsubscript{2} emission inventory without a prior emission estimate, instead using inventories of CO, NO\textsubscript{x}, and SO\textsubscript{2}. Similarly, Peischl et al. (2013) made use of CO and CO\textsubscript{2} inventories to help quantifying sources of CH\textsubscript{4} in the Los Angeles basin. The ability to measure multiple species has proved useful, also in remote sensing. For example, Pandey et al. (2015) made use of simultaneously retrieved CO\textsubscript{2} and CH\textsubscript{4} total column to reduce scattering effect. Further examples of studies making use of co-emitted species can be found in atmospheric chemistry (Konovalov et al., 2014; Berezin et al., 2013; Pison et al., 2009). More focused on exploiting inter-species correlation to reduce uncertainty in Bayesian inversion, Palmer et al. (2006) made use of CO\textsubscript{2}–CO correlations to improve inversion using data from the TRACE-P aircraft mission, while Wang et al. (2009) employed a similar method using satellite data, obtaining a reduction in the flux error of a CO\textsubscript{2} inversion.

So far the lion’s share of the studies investigating atmospheric inversions make use of both continuous in situ and flask measurements from ground-based observational networks of tall towers (e.g. Kadygrov et al., 2015; Sasa\textsubscript{k}awa et al., 2010). However, as profiles collected from aircraft easily exceed the height of towers, airborne data may also offer an interesting option. This alternative was tested in some recent studies that made use of aircraft profiles alone or in combination with other data sources (e.g. Brioude et al., 2013; Gourdi et al., 2012). Methods to maximize the cost-effectiveness of airborne data are the use of unmanned aircraft (drones) and commercial airliners. The latter, in particular, allow for collecting data on a regular basis without requiring a particularly small or light sensor. The most important projects making use of commercial airliners are CONTRAIL (Comprehensive Observation Network for Trace Gases; Machida et al., 2008) and MOZART-IAGOS (Measurements of Ozone and water vapour by in-service Airbus aircraft/In-service Aircraft for a Global Observing System; Marenco et al., 1998; Petzold et al., 2015). Both projects have been running for more than 2 decades and have produced extensive datasets that have proven to be important in the fields of atmospheric modelling and satellite calibration and validation (Zbiden et al., 2013; Sawa et al., 2012). Regarding carbonaceous species, CONTRAIL has so far collected CO\textsubscript{2} mixing ratio measurement, while IAGOS is focused on CO. In the next years the IAGOS fleet will simultaneously provide CO, CO\textsubscript{2}, and CH\textsubscript{4} atmospheric concentration measurements (Filges et al., 2015), enabling the use of multi-species synergy in modelling applications. This synergy follows the fact that the collocated measurements share the same atmospheric transport and have partially correlated emission uncertainties.

This paper is focused on investigating the benefits of such a multi-species inversion on uncertainty reduction in comparison with a single-species inversion. To achieve this goal, we set up a synthetic experiment utilizing the measurement times and locations collected from the IAGOS projects in the year 2011. The present paper is intended to pave the way for future studies making use of multi-species IAGOS datasets when they become available. A receptor-oriented framework was set up to derive flux interactions between the atmosphere and the biosphere using IAGOS data. The modelling framework is composed of a Lagrangian particle dispersion model (LPDM, specifically the STILT model), a diagnostic biosphere–atmosphere exchange model (the VPRM model), gridded emission inventories, global tracer transport model output that provides the tracer boundary conditions for the regional domain, and a Bayesian inversion scheme. The present work is based on the modelling framework used in Boschetti et al. (2015) and builds upon that by adding other species, and using a formal Bayesian inversion. A multi-species inversion was carried out in order to exploit the correlations in uncertainties between CO\textsubscript{2}, CO, and CH\textsubscript{4}, specifically in their respective uncertainties in a priori anthropogenic emissions and in model representation error. The aim of this multi-species inversion is to provide better estimates of anthropogenic emissions, and, in the case of CO\textsubscript{2}, to better separate the biospheric from anthropogenic contributions. This paper is structured as follows: a short description of the different components of the modelling framework is given in Sect. 2;
in Sect. 3 we present and discuss our results; Sect. 4 gives the conclusions.

2 Material and methods

2.1 Modelling framework

Before describing the different models composing the modelling framework, we introduce some specific terminology to reduce ambiguity in Sect. 2.1.1–2.1.6. Quantities that can be observed are termed “species” or “trace gases”, corresponding in this case to total CO$_2$, CO, and CH$_4$. These three species are simulated using five “modelled species”, namely CO$_2$ from fossil fuels, CO$_2$ related to GEE (gross ecosystem exchange), CO$_2$ related to respiration, CO, and CH$_4$. Modelled species related to anthropogenic emissions are modelled as the sum of contributions from different “emission sectors” (Table 1) and “fuel types” (Table 2); for further differentiation, anthropogenic and biospheric contributions are split into monthly contributions. Simulated fluxes specific for different modelled species, emission sectors, fuel types, and months of the year are called “flux categories”. In this section, a brief description of the different models that make up the modelling framework is given. For more detailed information, see Boschetti et al. (2015).

2.1.1 Vertical profile input data

In this study the modelled profiles have identical structure to those collected from the IAGOS fleet of commercial airliners. More precisely, the spatial and temporal coordinates of different observations will be used as input for the modelling framework, whereas the observed values of atmospheric mixing ratios of CO and meteorological parameters themselves will play a role in calibrating the modelling framework.

Central for this work is the concept of the mixed layer (ML), the lower part of the troposphere in which trace gases are well mixed due to turbulent convection in the timescale of an hour or less, and in which the effect of regional surface–atmosphere fluxes is the strongest. As input to the inversion we use the enhancement of the species’ mixing ratio within the mixed layer relative to that in the free troposphere (FT), similar to the approach described in Boschetti et al. (2015). This mixed layer enhancement best reflects the influence of regional fluxes. To compute this, we take the average mixing ratio within the mixed layer and subtract the value taken at 2 km above the mixed layer top ($z_l$), i.e. well within the free troposphere. The $z_l$ is a very important parameter in atmospheric modelling, and accounts for most of the transport uncertainty in the vertical domain. In fact, when assuming that the mixed layer is the part of the troposphere in which trace gases are well mixed due to turbulent convection, given a certain amount of trace gas in the ML, its mixing ratio will depend strongly on its depth $z_l$. More precisely, even if the model has correctly reproduced the amount of trace gas in the real mixed layer, if the modelled $z_l$ is lower (higher) than the actual one, then the simulated ML mixing ratio will be higher (lower) than it actually should be. In the present study, modelled $z_l$ is corrected according to Boschetti et al. (2015, Sect. 2.2.1).

2.1.2 Transport–flux coupling

The modelling framework is composed of a regional transport model (STILT), the EDGAR (Emission Database for Global Atmospheric Research) emission inventory to model anthropogenic emissions, VPRM (Vegetation Photosynthesis and Respiration Model) to model emissions from the biosphere, and output from global transport models for lateral boundary conditions for the different modelled species. The expressions “anthropogenic emissions” and “fossil fuel emissions” are considered synonymous in this paper and are used to indicate the sum of fossil fuel and biofuel emissions, without including contributions from LULUCF (Land Use, Land-Use Change and Forestry).

For regional transport we make use of the LPDM STILT (Stochastic Time-Inverted Lagrangian Transport; Lin et al., 2003) to derive the sensitivity of the atmospheric mixing ratio measurement to upstream surface–atmosphere fluxes, so-called “footprints”. Briefly, for each measurement location and time (also called receptor point), the model releases an ensemble of virtual particles that are driven back in time using wind fields from ECMWF and turbulence as stochastic process; the residence time within the lower half of the mixed layer is used to determine the potential contribution from surface fluxes, and the cumulative sum of these contributions determines the footprint that identifies the part of the domain with a certain influence on a single receptor point. To represent the mixed layer enhancements, the footprints for receptors within the boundary layer are averaged, and the footprint for the free tropospheric receptor is subtracted from this, resulting in a footprint for the mixed layer enhancements. This footprint is then matrix-multiplied with an emission map from an emission inventory, resulting in a simulated mixing ratio enhancement corresponding to the regional contribution at the measurement location.

A detailed description of STILT is given in Lin et al. (2003) and Gerbig et al. (2003). We use STILT coupled with emission models for both anthropogenic (EDGAR) and biosphere (VPRM) fluxes on a regional domain that covers most of Europe (33 to 72° N, −15 to 35° E) with a spatial resolution of 1/8° for latitude and 1/12° for longitude, roughly corresponding to 10 km. The MACC reanalysis (Inness et al., 2013, downloaded from http://www.ecmwf.int, last access: 23 June 2016) was used for lateral boundary conditions for CO mixing ratios, whereas for CO$_2$ and CH$_4$ we used output from the Jena CarboScope (Rödenbeck et al., 2003; CO$_2$ data available from www.bgc-jena.mpg.de/CarboScope/, last access: 10 February 2016), which are based on forward simulations of global-inversion optimized fluxes with the TM3 model.
Table 1. Specific emission sectors accounted for in the state vector and aggregated categories as used in Fig. 8.

<table>
<thead>
<tr>
<th>Adj IPCC</th>
<th>Description</th>
<th>Aggregated</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 1a1a</td>
<td>power generation</td>
<td>energy</td>
</tr>
<tr>
<td>2 1a1bcr</td>
<td>other transformation non-energy use</td>
<td>energy</td>
</tr>
<tr>
<td>3 1b1</td>
<td>solid fuel production</td>
<td>energy</td>
</tr>
<tr>
<td>4 1b2abc</td>
<td>gas flaring</td>
<td>energy</td>
</tr>
<tr>
<td>5 1b2ac</td>
<td>oil prod., distribution, and flaring</td>
<td>energy</td>
</tr>
<tr>
<td>6 1b2b</td>
<td>gas production and distribution</td>
<td>energy</td>
</tr>
<tr>
<td>7 1a3a+1c1</td>
<td>international and domestic aviation</td>
<td>transport</td>
</tr>
<tr>
<td>8 1a3b</td>
<td>road transport</td>
<td>transport</td>
</tr>
<tr>
<td>9 1a3ce</td>
<td>non-road ground transport</td>
<td>transport</td>
</tr>
<tr>
<td>10 1a3d+1c2</td>
<td>inland waterways and shipping</td>
<td>transport</td>
</tr>
<tr>
<td>11 1a2+6cd</td>
<td>industrial combustion (non-power)</td>
<td>industry</td>
</tr>
<tr>
<td>12 2a</td>
<td>cement and lime production</td>
<td>industry</td>
</tr>
<tr>
<td>13 2befg +3</td>
<td>chemical industry and solvents</td>
<td>industry</td>
</tr>
<tr>
<td>14 2c</td>
<td>metal industry emission</td>
<td>industry</td>
</tr>
<tr>
<td>15 1a4</td>
<td>buildings</td>
<td>buildings</td>
</tr>
<tr>
<td>16 4a</td>
<td>enteric fermentation in agriculture</td>
<td>agriculture</td>
</tr>
<tr>
<td>17 4b</td>
<td>manure management</td>
<td>agriculture</td>
</tr>
<tr>
<td>18 4c</td>
<td>rice cultivation</td>
<td>agriculture</td>
</tr>
<tr>
<td>19 4f</td>
<td>agricultural waste burning</td>
<td>agriculture</td>
</tr>
<tr>
<td>20 6a</td>
<td>solid waste disposal in landfills</td>
<td>waste</td>
</tr>
<tr>
<td>21 6b</td>
<td>wastewater treatment</td>
<td>waste</td>
</tr>
<tr>
<td>22 7a</td>
<td>fossil fuel fires</td>
<td>FF_fuels</td>
</tr>
</tbody>
</table>

Table 2. Specific fuel types accounted for in the state vector and aggregated categories as used in Fig. 8.

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Aggregated fuel type</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 brown coal</td>
<td>coal</td>
</tr>
<tr>
<td>2 hard coal</td>
<td>coal</td>
</tr>
<tr>
<td>3 peat</td>
<td>coal</td>
</tr>
<tr>
<td>4 gas derivatives</td>
<td>gas</td>
</tr>
<tr>
<td>5 natural gas</td>
<td>gas</td>
</tr>
<tr>
<td>6 heavy oil</td>
<td>oil</td>
</tr>
<tr>
<td>7 light oil</td>
<td>oil</td>
</tr>
<tr>
<td>8 solid waste</td>
<td>waste</td>
</tr>
<tr>
<td>9 venting and flaring</td>
<td>oil</td>
</tr>
<tr>
<td>10 other*</td>
<td>other</td>
</tr>
<tr>
<td>11 gas biofuels</td>
<td>bio</td>
</tr>
<tr>
<td>12 liquid biofuels</td>
<td>bio</td>
</tr>
<tr>
<td>13 solid biofuels</td>
<td>bio</td>
</tr>
</tbody>
</table>

* The category “other” is derived by summing the contribution from those processes in which it is difficult to establish the specific fuel responsible for the emissions.

For fossil fuel emissions, we use a model based on the EDGAR v4.3.1 emission inventory (European Commission, 2016) modified following the same approach taken for COFFEE (CO$_2$ release and Oxygen uptake from Fossil Fuel Emission Estimate; Steinbach et al., 2011; Vardag et al., 2015). More precisely, to obtain hourly resolved emissions from the original EDGAR annual fluxes for different emission categories we add specific temporal activity factors (Denier van der Gon et al., 2011) to account for differences in emissions due to seasonal, weekly, and daily cycles. In addition, the different emission categories are further split into contributions from different fuel types from British Petroleum's Statistical Review of World Energy 2014 (BP, 2014). The World Energy Outlook from IEA as alternative source of information was not chosen, as the report from BP was available earlier (April 2015 vs. November of the following year). This allows for taking into account changes in emissions between different years. Such an emission model provides hourly resolved fluxes for each fossil fuel flux category with a spatial resolution of roughly 10 km on our regional European domain. For each of the three anthropogenic modelled species (CO$_2$, CO, and CH$_4$), different emission maps are used as input. Temporal profiles are then applied to these sector- and fuel-specific emission maps. To also take into account the contribution from the biosphere we use VPRM. VPRM simulates realistic patterns at small spatial (10 km x 10 km) and temporal (hourly) scales and is used here to provide the a priori fluxes for biosphere–atmosphere exchange of CO$_2$. This model is described in detail in Mahadevan et al. (2008).
STILT transport is driven by meteorological fields from the ECMWF IFS (12 h forecasts twice daily at 3-hourly temporal resolution), which have a spatial resolution of 0.25° with 61 vertical levels. In the following, we will refer to the STILT/EDGAR/VPRM/MACC/TM3 combination of transport, simulated fluxes and advected boundary conditions as merely “STILT” for simplicity.

2.1.3 Bayesian inversion

Atmospheric inversions provide an estimate of the distribution of sources and sinks over the domain’s surface from available concentration measurements (top-down approach). This can be formalized in the following linear relation:

\[ y = K\lambda + \varepsilon, \]

where the \( y \) vector contains the \( n \) observations, and \( K \) is the Jacobian matrix that relates the observations with the state vector \( \lambda \). In the present study the focus will be on surface–atmosphere gas exchanges due to biospheric processes and anthropogenic emissions. So the observations are trace gas mixing ratios at different times and locations, \( K \) is the product of a transport operator \( H \) that maps flux sensitivities at different times and locations with a set of gridded fluxes \( F \) for the categories of interest, while the state vector \( \lambda \) contains the \( m \) scaling factors for the flux categories of interest. \( H \) has \( n \) rows and a number of columns equal to \( h = N_x \times N_y \times N_t \times N_s \) respectively the number of pixels in the emission model along the \( x \) and \( y \) axes, the number of (hourly) simulations in the whole year of interest, and the number of state vector elements, resulting in a huge matrix. As the matrix \( F \) describes the different simulated gridded fluxes, it is comparably large and has \( h \) rows and \( m \) columns.

By considering \( K \) as the result of the product of these two large matrices, it is possible to limit its dimensions to only \( n \) rows and \( m \) columns; this allows for simplifying the critical task of relating observation with simulated fluxes of the categories of interest. The state vector accounts for specific categories of interest. The state vector \( \lambda \) is the result of the product of these two matrices, the number of state vector elements, resulting in a huge matrix. The error covariance matrix of the optimal posterior state (the posterior uncertainty) is given by

\[ S_{post} = (K^T S_{\varepsilon}^{-1} K + S_{prior}^{-1})^{-1}. \]

Note that this quantity depends on neither the prior fluxes nor the measured mixing ratios, but only on their respective uncertainties and on the transport matrix \( K \). In this study, the inverse of the matrices was calculated using the R function “solve” from the base package of R version 3.0.0 (http://www.r-project.org/, last access: 30 April 2013).

The targeted quantities of this study are the aggregated emissions over a specific area at a specific timescale (e.g. month); those quantities can be derived from the prior and posterior state through a spatiotemporal aggregation operator \( \Lambda \) that allows for the conversion of scaling factors into physically representative quantities. As the pseudo-observations are clustered around a single location (Frankfurt), it is very likely that the fluxes over the whole European domain are not constrained. Therefore, as a spatial aggregation scale we chose an area from which fluxes have a significant contribution to the observations made at Frankfurt. For this we compute the temporally accumulated footprint values (cf. Sect. 2.1.2) for the whole year 2011, and select those spatial pixels that correspond to 50 % of the total (spatially integrated) footprint (Fig. 1). Note that by using this aggregation
scale we assume perfectly known distribution within a given flux category that can result in aggregation error, especially with respect to biogenic fluxes, that are not as well known as anthropogenic fluxes. However, the chosen domain of aggregation is quite small, and the total anthropogenic fluxes are divided according to species, emission categories, fuel types, and months. This results in 69 degrees of freedom per month for each anthropogenic species and 10 degrees of freedom per month for the biospheric fluxes; for this reason we expect the aggregation error not to be a particularly important source of uncertainty. The prior and posterior uncertainty of these targeted quantities ($\sigma_{\text{prior}}$ and $\sigma_{\text{post}}$) is obtained by applying the aggregation operator to the respective uncertainty covariances:

$$\sigma_{\text{prior}} = \sqrt{A^T S_{\text{prior}} A} \quad \text{and} \quad \sigma_{\text{post}} = \sqrt{A^T S_{\text{post}} A}. \quad (5)$$

Different versions of the aggregation operator were created for this: emissions categories are aggregated according to different fuel types (coal, oil, gas, bio, waste, and other) and according to emission sectors (energy, transport, industry, buildings, agriculture, waste, and fossil fuel fires). Note that only these aggregated fluxes are optimized, not the individual gridded fluxes of the emission inventories.

To quantitatively assess the information provided by the inversion, the reduction of uncertainty in the posterior compared to the prior estimate is a useful measure. The uncertainty reduction (UR) is defined as

$$\text{UR} = 1 - \frac{\sigma_{\text{post}}}{\sigma_{\text{prior}}}. \quad (6)$$

The uncertainty reduction ranges from 0 (posterior as large as the prior uncertainty) to 1 (posterior negligible compared to the prior uncertainty).

### 2.1.4 Prior error structure

As in this study a multi-species inversion with CO, CO$_2$, and CH$_4$ is envisioned, we have the chance to exploit the correlations in the uncertainties of the different trace gases related to both a priori fluxes and model–data mismatch. This is particularly true for CO and CO$_2$ because they share a larger part of the emission sources, which implies correlations in the respective uncertainties. In the multi-species inversion, such information is stored in the areas of the error covariance matrices that describe covariance between different modelled species (off-diagonal “blocks” in Fig. 2b for $S_{\text{prior}}$ and Fig. 3b for $S_{\epsilon}$). In the single-species inversions, said covariance is set to zero, corresponding to a situation where the different species are completely independent of one another. Conversely, the measurement uncertainty is stored in the main diagonal of the $S_{\epsilon}$ (Fig. 3d).

We used a single year (2011) dataset restricted to the vertical profiles centred at the Frankfurt airport (FRA) and restricted to daytime during well-mixed atmospheric conditions (10:30 to 17:30 CET). The dataset contains 1098
pseudo-observations, 366 for each of the three observable species, whereas the state vector contains the scaling factors for 2604 flux categories, each equal to one in the prior.

The prior error covariance matrix can be expressed as follows:

$$S_{\text{prior}} = C_{\text{prior}} \rho_{\text{prior}}, \quad (7)$$

where $C_{\text{prior}}$ is the prior error correlation matrix (Fig. 2a) and $\rho_{\text{prior}}$ is a prior rescaling matrix described in Sect. 2.1.5 (Fig. 4a). First we describe how $C_{\text{prior}}$ is generated. The prior error correlation matrix is a square matrix of rank 2604, reflecting the length of the state vector, and results from the product of three components (Fig. 2b, c, and d) accounting for correlations between flux categories according to the modelled species, emission sectors, and fuel types respectively. In four different instances, a correlation of 0.7 is applied:

1. between different anthropogenic modelled species;
2. between GEE and respiration;
3. between different emission sectors;
4. between different fuel types.

Such a correlation implies that the explained variance for each constraint, everything else being equal, is roughly 50 % (0.7 to the square equals 0.49), with the rest remaining independent. In addition, the correlation between fossil-fuel-related and biosphere-related scaling factors is zero, and the same holds for fluxes of different months, indicating complete independence from one another. In this study, we assume a certain annual total domain-wide flux uncertainty, and then break it down by sectors, fuels, and months by inflating the error. By assuming no correlation between different months we ensure maximum flexibility in the system to retrieve month-to-month changes based on the observations. We assume correlation between months is possible, but it has not been investigated here. It is unclear how good the seasonal variation in emissions from the inventories actually is; so in order to not rely too much on these we chose zero correlation. Investigating the effects of different correlation set-ups for the seasonal cycle could be the focus of future research.

### 2.1.5 Prior error scaling

After having specified the prior error correlation matrix $C_{\text{prior}}$, we now describe how we rescale it to obtain $S_{\text{prior}}$; for this task we rewrite Eq. (7) as

$$S_{\text{prior}} = C_{\text{prior}} \rho_{\text{prior}} =$$

$$= \begin{pmatrix}
C_{11} & C_{12} & C_{13} & 0 \\
C_{21} & C_{22} & C_{23} & 0 \\
C_{31} & C_{32} & C_{33} & 0 \\
0 & 0 & 0 & C_{\text{bio}}
\end{pmatrix}$$

Table 3. Relative uncertainty of the prior fluxes aggregated domain-wide and annually for the different cases.

<table>
<thead>
<tr>
<th></th>
<th>CO 2</th>
<th>CO</th>
<th>CH 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>20 %</td>
<td>50 %</td>
<td>50 %</td>
</tr>
<tr>
<td>Case 2</td>
<td>10 %</td>
<td>50 %</td>
<td>50 %</td>
</tr>
<tr>
<td>Case 3</td>
<td>10 %</td>
<td>25 %</td>
<td>25 %</td>
</tr>
</tbody>
</table>

$$\begin{pmatrix}
1/\rho_1 & 1/\rho_2 & 1/\rho_3 & 0 \\
1/\rho_2 & 1/\rho_3 & 0 & 0 \\
1/\rho_3 & 0 & 0 & 0 \\
0 & 0 & 0 & \rho_{\text{bio}}^2
\end{pmatrix}, \quad (8)$$

where each $C_{ij}$ is a subset of the fossil fuel part of $C_{\text{prior}}$ (block) as shown in Fig. 2, and each $\rho_i$ is defined as

$$\rho_i = \sqrt{\sum_j A_{ij}' A_{i}'}, \quad (9)$$

where $A'$ is the aggregation operator for annual fluxes over the full domain, and $\epsilon_j$ is the corresponding relative prior uncertainty, assuming the values specified in Table 3 for different cases. Case 1 is considered as the default case, with prior uncertainty values conservatively assumed to be 20 % for CO2, and 50 % for CO and CH4. Conversely, $C_{\text{bio}}$ covers the biospheric part of $C_{\text{prior}}$, and for $\sum_j A_{ij}' \epsilon_j$ for $\rho_{\text{bio}}$ we use a prior uncertainty of 0.54 GtC y⁻¹, as derived in Kountouris et al. (2018) for the VPRM model. The biospheric part of the prior error covariance matrix assumes no correlation with the fossil fuel species.

The posterior of each Bayesian inversion depends on its specific prior. As the multi- and single-species inversions have different prior uncertainty structures, the uncertainty reduction for targeted quantities cannot be directly compared (Eq. 4). To be able to compare the two inversions, we require that the a priori aggregated uncertainty of the targeted quantities remains the same, and distribute it differently each time; the prior rescaling matrix $\rho_{\text{prior}}$ is needed for this task. The benefits were tested for observations taken in different months and for three different error structures in the prior uncertainty. As a priori aggregated uncertainty we use a percentage of the aggregated modelled emissions for fossil fuels across the whole year. Table 3 shows the percentage values used for different cases.

### 2.1.6 Model–data mismatch error structure

In an atmospheric inversion, the model–data mismatch from every uncertainty source (such as measurement uncertainty, transport model uncertainty, spatial representation error due to limited model resolution, and boundary condition inaccuracies) needs to be taken into account. In our inversion
scheme, we parameterize both the transport model uncertainty and the measurement uncertainty, with the latter playing a minor role. The model–data mismatch covariance matrix ($S_t$) is constructed according to the following equation:

$$S_t = C_sC_t\epsilon_{\text{tran}}^2 + \epsilon_{\text{meas}}^2,$$

where $C_s$ accounts for correlations between different observed species (Fig. 3b), $C_t$ accounts for the temporal correlation (Fig. 3c), $\epsilon_{\text{tran}}$ is the total transport error, and $\epsilon_{\text{meas}}^2$ accounts for all of the non-transport-related errors like spatial representation error and lateral boundary conditions (Fig. 3d).

The assumed measurement uncertainty is 1 ppm for CO$_2$, 20 ppb for CO, and 20 ppb for CH$_4$, while $\epsilon_{\text{tran}}$ is time dependent and assumed to be proportional to the modelled enhancement due to regional fluxes. The assumed measurement uncertainty is higher than the expected instrument precision because it also includes in addition the uncertainties related to spatial representation and lateral boundary conditions. $\epsilon_{\text{tran}}$ is characterized as follows by different components in the vertical and horizontal domain:

$$\epsilon_{\text{tran}} = \text{enh}\sqrt{\epsilon_{\text{tran}_h}^2 + \epsilon_{\text{tran}_v}^2},$$

where enh indicates the modelled enhancement, and both the horizontal transport error $\epsilon_{\text{tran}_h}$ and the vertical transport error $\epsilon_{\text{tran}_v}$ are characterized as percentage error; $\epsilon_{\text{tran}_h}$ is assumed to be 50%, while $\epsilon_{\text{tran}_v}$ is a profile-specific relative error with a mean value of about 10%. The vertical transport error accounts for the fact that the shallower the mixed layer is, the more difficult it is to model the atmosphere. We assume that after $z_i$ correction the remaining error is of the order of 50 m (related to the vertical resolution of the profile data), so the relative error $\epsilon_{\text{tran}_v}$ is assumed to be the ratio of 50 m to the modelled $z_i$; in this way we obtain an error that gets larger the shallower the mixed layer is. For the horizontal component, an uncertainty of 50% is a conservative estimate based on Lin and Gerbig (2005), where the horizontal transport error is found to be 5.9 ppm for CO$_2$. This, combined with about 10 ppm of drawdown in the mixed layer relative to the free troposphere, gives something like 50% error in the regional flux signal. The vertical component is so much smaller in percentage since the simulated mixing ratios are already corrected for mismatch between modelled and observed $z_i$.

In the multi-species inversion, the transport error correlation across species is 0.7 (Fig. 3b), while in the single-species inversion this is set to zero. Time correlation is assumed to decay exponentially with an exponential constant of 12 h. The between-species correlation for model–data mismatch related to transport uncertainty reflects the fact that species are partially co-emitted and share the same atmospheric transport (and its related uncertainty).

### 2.2 Synthetic experiment

#### Pseudo-data generation

As explained in the introduction, in situ measurements are not available for all of the three trace gases of interest, but only for CO. For this reason this paper aims to evaluate the benefits of a multi-species inversion over a corresponding single-species inversion by performing a synthetic experiment, using pseudo-observations derived by perturbation of the model outputs based on a priori state vector values. More precisely, the pseudo-observation vector is obtained by matrix multiplication between the Jacobian matrix $K$ and what we assume to be the true state vector. The true state vector itself is obtained by using the sum of the prior state vector (all values equal to one) and a random realization of the prior error, truncated to avoid negative state vector values. In detail, the error realization is obtained by multiplying a randomly generated, normally distributed vector with the prior error covariance matrix. This ensures that such a realization has the same error correlation as the prior uncertainty. Where the
result of such matrix-vector product is negative, the same operation is performed recursively until all elements of the state vector are positive. This ensures that the difference between the true and prior state vector has the same error correlation structure as described by the prior error covariance matrix.

3 Results and discussion

Before evaluating the performance of the inversion scheme in reducing the uncertainty of the state space, a closer look at the ability of the modelling framework to reproduce the enhancements is necessary. Unfortunately, this can be done only for CO as actual measurements are not available for the other species. Figure 5 shows the mean daily enhancement of the three fossil fuel species for both observations and model outputs using prior emissions. A common feature of the three trace gases is that lower values tend to occur during summer time due to better mixing of the atmosphere. Conversely, enhancement values tend to be higher during winter, reflecting the more stratified atmosphere of the cold months.

In Fig. 5 the modelled CO plot was multiplied by a factor of 2.8, corresponding to the mean ratio between observed and modelled CO enhancements, similar to what was found in Boschetti et al. (2015). Mixing ratio values are highly variable, but the model provides a good indication of the temporal variation of the ML enhancement; the squared correlation coefficient between observed and modelled CO enhancements is 0.62, while the standard deviation of corrected model and observation residuals is 85 ppb; note that by not accounting for the \( z_j \) correction, such values would be 0.56 and 87 ppb respectively. The median of the mixing ratio enhancement for the three trace gases is 2.8 ppm for CO\(_2\), 18.6 ppb for CO, and 26.6 ppb for CH\(_4\). For CO\(_2\) this seasonal difference is enhanced due to the simultaneous presence of both anthropogenic and biogenic emissions. During summer, values are slightly negative due to strong photosynthesis fluxes from growing vegetation from the active combined with deeper vertical mixing. Negative values arise in 31% of the cases predominantly during the warmer months, implying that during the growing period uptake by photosynthesis dominates over release from combustion and respiration. Both CO and CH\(_4\) experience higher values during winter due to the shallow mixed layer usually associated with cold temperatures, and lower values during summer as higher temperatures cause the mixed layer to reach higher altitudes; differences related to seasonal domestic heating and transportation may also play a role. In addition, enhancement for both species is occasionally negative, most likely owing to advection of polluted air masses in the free troposphere. An alternative explanation is that strong winds at lower heights can disperse the emissions in the boundary layer and create a situation in which the mixing ratio in the FT is higher than in the ML.

Figure 6 shows the prior and posterior error covariance matrices for the base multi-species inversion. Note that CO\(_2\) from anthropogenic emissions is assumed to be independent from biogenic emissions; therefore prior error correlation between these categories is zero. The posterior error covariance matrix for the multi-species inversion (Fig. 6b) shows lower values corresponding to an average uncertainty reduction of 23% across all state vector elements, while the posterior error covariance matrix for the single-species inversion (not shown) is characterized by a mean uncertainty reduction of 20%. This result implies that the multi-species inversion improves the uncertainty reduction by roughly 15%. Negative values in the posterior error correlation matrix are to be expected because different categories are bound together by correlations and therefore are not free to vary independently.

Figures 7 and 8 show a priori, a posteriori, and “true” fluxes related to different aggregated fuel types and to different emission categories as described in Tables 1 and 2 for the months of July and December. Figure 8 also shows the biospheric contribution (as absolute values) scaled down by a factor of 10. As is to be expected, the biospheric contributions show strong differences according to the seasonal cycle, while anthropogenic emissions remain rather stable. However, it is worth pointing out that while the fossil fuel prior is similar for both months, the assumed truth can be rather different due the random assignment of the prior error realization. In most cases, the posterior adapts and is therefore closer to the truth than the prior; the posterior uncertainty is also visibly reduced, as expected. Regarding the different tracers, CO\(_2\) and CO show a somewhat similar pattern indicating a partial overlap in dominating emission categories while CH\(_4\) is dominated by different contributions in both fuel types and emission categories.

Our modelling framework is currently not well suited to account for unreported sources of CH\(_4\) due to the lack of information about natural gas and oil production operations, or from recent and old mining areas. Many recent studies have discussed the problem, mainly referring to shale basins exploited via hydraulic fracturing in the USA (e.g. Kort et al., 2016; Karion et al., 2015; Lyon et al., 2015). For example, Karion et al. (2015) concludes that EDGAR underesti-
Figure 5. Mean daily enhancement of mixed layer vs. free tropospheric mole fractions. Modelled mixing ratios are shown as black lines, while the observed CO is shown as a blue line. Note that the modelled values for CO have been multiplied by a factor of 2.8, corresponding to the mean ratio between observed and modelled CO enhancements, to match the observed values.

Figure 6. Prior error covariance matrix (a) and corresponding posterior error covariance matrix (b).

In general, the absence of some emission sources in an inventory is equivalent to the assumption of having point sources not included in the emission map, but still contributing to the measurements. The inversion scheme would typically react to this by assigning such point sources in some other sectors to another fuel type. As a result, the posterior enhancements would be biased low in proximity of those point sources, and (slightly) biased high for influences from other regions with the same sector or fuel type. This issue should definitely be considered in future study making use of actual CO, CO$_2$, and CH$_4$ observations from IAGOS but has limited effects on this paper, as our main focus is on the benefits of inter-species correlation on the posterior uncertainty in a synthetic experiment.

Note that our modelling framework does not allow for simulating CO biogenic fluxes during the growing season. Warm days in summer correspond to large amount of biogenic volatile organic compounds (VOCs) being emitted from vegetation, producing CO at non-negligible levels. According to Hudman et al. (2008), anthropogenic emissions account for only 31% of CO emissions in the USA during summer. Conversely, according to estimates from EDGAR, CO an-
Dominant fuels (Fig. 7) for CO\textsubscript{2} are coal, gas, and oil, whose prior fluxes (pseudo-data) have a magnitude of 6–11 Megatons of carbon per year (MtC month\textsuperscript{−1}) in July and 8–14 MtC month\textsuperscript{−1} in December. For CO the most important emission sector is heating of buildings during winter, contributing a flux of 0.19 MtC month\textsuperscript{−1}. Contributions from industry and transport dominate during summer, with magnitudes of 0.04 and 0.05 MtC month\textsuperscript{−1}, respectively. The dominant fuel for CO is biofuel with 0.19 MtC month\textsuperscript{−1} emissions during winter. The secondary industrial and transport contributions originate in summer from oil and biofuels with a magnitude of 0.06–0.08 MtC month\textsuperscript{−1} and from agricultural waste burning with a magnitude of 0.06–0.11 MtC month\textsuperscript{−1}.

Contrary to CO\textsubscript{2} and CO, CH\textsubscript{4} is determined by non-combustion sectors, more specifically by a contribution of 0.15 MtC month\textsuperscript{−1} flux from agriculture (manure management and rice cultivation) in July with secondary contributions from waste and energy with a magnitude of roughly 0.06–0.08 MtC month\textsuperscript{−1} in both July and December. Other non-combustion sectors, in particular wastewater treatment and landfills contribute to a total of 0.16–0.24 MtC month\textsuperscript{−1} of emissions. These non-combustion sectors contribute to less than 20 % of total CO\textsubscript{2} emissions, with 1.13 MtC month\textsuperscript{−1} from the cement and lime industry and less than 20 % of the total CO emissions (0.03 MtC month\textsuperscript{−1} from the metal industry).

The contribution to CO\textsubscript{2} from biospheric primary production (a sink for atmospheric CO\textsubscript{2}) is about 100 MtC month\textsuperscript{−1} in July, which drops to almost zero in December, while respiration values are 50 MtC month\textsuperscript{−1} in July and roughly 15 MtC month\textsuperscript{−1} in December.

As further assessment of the inversion performance, we tested the ability of the inversion scheme to capture the truth compared with a perturbed version of the prior. Such perturbed version is obtained by adding a random distribution with mean and standard deviation equal one to the prior state space, similar to how the truth is obtained. For each simulated species we calculated the total annual fluxes for prior, posterior, truth, and perturbed prior. From these total fluxes we then derive the overall residual between prior and truth, posterior and truth, and perturbed prior and truth. It is clear from Table 4 that while the overall bias between posterior and truth is lower than the prior–truth bias, the bias between perturbed prior and truth is much higher, implying that the performance of the inversion is not an artifact of the pseudo-data generation. In addition, it was found that the truth–prior bias of the multi-species inversion is mostly slightly lower compared to the single-species inversion. The difference is between −2.2 and 7.6 %, according to the simulated species, with an overall value of 0.3 %.

Improper characterization of the error correlation may result in systematic bias in the posterior estimate. As mentioned in Sect. 2.1.6, inter-species correlation, the correlation between different fuel types and the correlation between different emission sectors in S\textsubscript{prior}, is assumed equal to 0.7 (Sect. 2.1.4). To assess how well the system will reproduce the true fluxes with incorrectly specified correlations, a series of experiments was performed in which the inter-species correlation in S\textsubscript{prior} remains equal to 0.7, while the three correlation coefficients in S\textsubscript{prior} assume different values ranging from 0.1 to 0.9. Table 5 shows the residuals between total annual posterior fluxes and total annual true fluxes for the five simulated species, derived similarly as for Table 4. We found that for all species the uncertainty reduction increases with correlation. More precisely, from correlation 0.1 to 0.9, the annual uncertainty reduction for anthropogenic CO\textsubscript{2} increases from 26.6 to 51.7 %, while the increase is lower for GEE (from 72.4 to 73.1 %) and respiration (from 39.3 to 41.3 %) because the biospheric fluxes are independent from other species. For CO, the uncertainty reduction increases from 60.7 (with correlation 0.1) to 66.4 % (with correlation 0.9). The annual uncertainty reduction for CH\textsubscript{4} increases from 60.5 to 67.5 %.
Table 4. Overall bias for different species between the prior and both posterior and perturbed prior. The percentage values in parentheses are the corresponding prior–truth bias.

<table>
<thead>
<tr>
<th>Species</th>
<th>Prior-truth (MtC y(^{-1}))</th>
<th>Posterior-truth (MtC y(^{-1}))</th>
<th>Pert. prior-truth (MtC y(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO(_2) FF</td>
<td>-14.2 (-111%)</td>
<td>1.5 (+111%)</td>
<td>-8.8 (-38%)</td>
</tr>
<tr>
<td>CO</td>
<td>-0.95 (-69%)</td>
<td>-0.29 (-69%)</td>
<td>-1.08 (+13%)</td>
</tr>
<tr>
<td>CH(_4)</td>
<td>0.36 (68%)</td>
<td>0.11 (-68%)</td>
<td>0.84 (+133%)</td>
</tr>
<tr>
<td>GEE</td>
<td>-81.8 (-78%)</td>
<td>-17.9 (-78%)</td>
<td>-116.8 (+43%)</td>
</tr>
<tr>
<td>Respiration</td>
<td>39.5 (-48%)</td>
<td>20.6 (-48%)</td>
<td>62.2 (+58%)</td>
</tr>
</tbody>
</table>

Figure 9. Comparison between prior and posterior monthly uncertainties for the five tracers. The posterior uncertainty is plotted for both the multi-species inversion, accounting for inter-species correlations, and the single-species inversion, in which all of the species are independent. Both prior and posterior uncertainty are expressed in physical units. The spike in the prior methane uncertainty estimate for the month of March depends on the emission inventory and is related to the cycle of agricultural activities.
correlations shown in Fig. 10 does not depend on different manifestations of the true fluxes, but only on the posterior uncertainty of the multi- and single-species inversions.

All of the species experience a reduction in the posterior uncertainty ratio due to the addition of inter-species correlation; said reduction is up to 20% for fossil fuel CO$_2$ and up to 10% for the other species; in addition, anthropogenic CO$_2$ is more sensitive to the prior relative error values than CO and CH$_4$. As the uncertainty of GEE and respiration is not modified, they show little to no variation for different cases (Fig. 10). There is a dependence of the benefit of the multi-species inversion over the single-species inversion on the prior uncertainty values (differences between Cases 1–3), with the largest difference for fossil fuel emissions of CO$_2$. Interestingly for Case 2 with reduced prior uncertainty for fossil fuel CO$_2$ emissions the benefit nearly doubles over the default case (Case 1). Also reducing the prior uncertainties of CO and CH$_4$ emissions (Case 3) more or less compensates for this increase in benefit. The reason for both of these results is probably to be found in Eq. (8). In fact, changing the prior uncertainty in CO$_2$ emissions means to also change the off-diagonal blocks linking the different species together. However, by reducing the anthropogenic CO$_2$ uncertainty from 20 to 10% (Case 2), the diagonal block for CO$_2$ in the prior uncertainty changes by a factor of 4, while the off-diagonal blocks change only by a factor of 2. This effectively ties the emissions of CO$_2$ tighter to the emissions of the other species, resulting in greater benefit from a multi-species inversion over a single-species inversion. Conversely, when all prior uncertainties are reduced by a factor of 2 (Case 3), both diagonal and off-diagonal blocks are reduced by a factor of 4. This explains why Case 1 and Case 3 show similar benefit values. Note that the assumed prior uncertainties for the default case (Case 1) are quite conservative; therefore lower uncertainties were chosen for Cases 2 and 3. While the absolute benefit of adding inter-species correlation is not a game-changer, it is worth pointing out that such improvement also comes with only slightly greater computational effort than multiple independent single-species inversions.

### Table 5. Residuals between total annual posterior fluxes (post) and total annual true fluxes (truth) for the five simulated species (in MtC yr$^{-1}$) and different inter-species correlation values in the prior error covariance matrix (first column). The corresponding posterior uncertainty was added for each post–truth value.

<table>
<thead>
<tr>
<th>Correlation</th>
<th>Post–truth CO$_2$ FF</th>
<th>Post–truth CO</th>
<th>Post–truth CH$_4$</th>
<th>Post–truth GEE</th>
<th>Post–truth respiration</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>$-6.3 \pm 16.4$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$-0.1 \pm 0.3$</td>
<td>$-18.5 \pm 23.6$</td>
<td>$-19.0 \pm 27.5$</td>
</tr>
<tr>
<td>0.2</td>
<td>$-4.4 \pm 16.1$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$0.0 \pm 0.3$</td>
<td>$-18.6 \pm 23.5$</td>
<td>$-19.2 \pm 27.4$</td>
</tr>
<tr>
<td>0.3</td>
<td>$-2.7 \pm 15.9$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$0.0 \pm 0.3$</td>
<td>$-18.6 \pm 23.4$</td>
<td>$-19.5 \pm 27.3$</td>
</tr>
<tr>
<td>0.4</td>
<td>$-1.3 \pm 15.6$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$0.0 \pm 0.3$</td>
<td>$-18.5 \pm 23.4$</td>
<td>$-19.7 \pm 27.3$</td>
</tr>
<tr>
<td>0.5</td>
<td>$-0.1 \pm 15.2$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$0.0 \pm 0.2$</td>
<td>$-18.4 \pm 23.3$</td>
<td>$-20.0 \pm 27.2$</td>
</tr>
<tr>
<td>0.6</td>
<td>$0.8 \pm 14.6$</td>
<td>$0.3 \pm 0.2$</td>
<td>$0.1 \pm 0.2$</td>
<td>$-18.2 \pm 23.2$</td>
<td>$-20.3 \pm 27.1$</td>
</tr>
<tr>
<td>0.7</td>
<td>$1.5 \pm 13.7$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$0.1 \pm 0.2$</td>
<td>$-17.9 \pm 23.2$</td>
<td>$-20.6 \pm 26.9$</td>
</tr>
<tr>
<td>0.8</td>
<td>$1.9 \pm 12.4$</td>
<td>$-0.3 \pm 0.2$</td>
<td>$0.2 \pm 0.2$</td>
<td>$-17.6 \pm 23.1$</td>
<td>$-20.9 \pm 26.8$</td>
</tr>
<tr>
<td>0.9</td>
<td>$1.5 \pm 10.4$</td>
<td>$-0.4 \pm 0.2$</td>
<td>$0.3 \pm 0.2$</td>
<td>$-17.3 \pm 23.0$</td>
<td>$-21.1 \pm 26.5$</td>
</tr>
</tbody>
</table>

In order to assess the contribution of inter-species correlation in the prior uncertainty vs. that of model–data mismatch uncertainty, Fig. 11 also shows the resulting posterior mismatch ratios for Case 1 (Table 3) from inversions only using prior or model–data mismatch correlation. For the anthropogenic component of CO$_2$, the greatest constraint is given by the prior correlation, while for GEE, respiration, and CH$_4$ the strongest contribution is from the model–data mismatch correlation. In the case of CO, the inter-species correlations for different components are dominant for different months of the year. What makes CO sensitive to different correla-
4 Conclusions

The present paper presents a synthetic experiment aiming to evaluate the effects of exploiting correlations between different trace gases in an atmospheric inversion. We quantitatively described the capability of the modelling framework to reproduce observations, the performance of the inversion scheme in reducing the uncertainty of the different trace gases, and the benefits of multi-species inversions compared to corresponding single-species inversions. We also describe a method to re-scale different prior uncertainty covariance matrices so that the corresponding posterior uncertainties are actually comparable.

Where possible, we compared model outputs with available observations. Such comparison, possible only for CO, showed a good degree of agreement between the model and observations with an overall correlation of roughly 0.75; modelled values for CO enhancement underestimate the observed ones by a factor of roughly 2.8, compatible with what was found in Boschetti (2015). It is found that posterior uncertainty is much lower than the prior for all of the five simulated species. The mean uncertainty reduction for CO2 emissions from fossil fuels is roughly 38 %, for GEE it is around 41 %, while for respiration it is roughly 44 %. For CO and CH4 the uncertainty reduction is about 63 and 67 % respectively. Finally, we described quantitatively the benefit of using multi-species inversions by exploiting correlations in different chemical species. It is found that considering correlations between different trace gases can reduce the posterior uncertainty by up to about 20 % for monthly fluxes. These
benefits are however dependent on the error structure of the prior uncertainty.

The present paper paves the way for future studies using simultaneous measurements of different trace gases. This will be especially important in the context of the upcoming routine measurements of CO$_2$, CO, and CH$_4$ vertical profiles within IAGOS. As IAGOS makes use of commercial airliners, such profiles will be collected in the vicinity of major international airports, and hence in the vicinity of major metropolitan areas, where many different human activities take place simultaneously. In such a context, any improvement in the constraint of atmospheric inversions will be particularly useful. A possible improvement in this analysis would be to evaluate the effects of different correlation factors specific to different pairs of anthropogenic species, fuels, and emission sectors.

Data availability. IAGOS and MOZAIC data for carbon monoxide mole fraction measurements are available at the IAGOS database under http://iagos.sedoo.fr/portal.html (last access: 14 November 2016).

Competing interests. The authors declare that they have no conflict of interest.

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