



## Age of Air as a diagnostic for transport time-scales in global models

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**Abstract.** This paper presents the first results of an age of air (AoA) inter-comparison of six global transport models. Following a protocol, three global circulation models and three chemistry transport models simulated five tracers with boundary conditions that grow linearly in time. This allows for an evaluation of the AoA and transport times associated with inter-hemispheric transport, vertical mixing in the troposphere, transport to and in the stratosphere, and transport of air masses between land and ocean. Since AoA is not a directly measurable quantity in the atmosphere, simulations of <sup>222</sup>Rn and SF<sub>6</sub> were also requested. We focus this first analysis on averages over the period 2000–2010, taken from longer simulations covering the 1988 – 2014 period. We find that two models, NIES and TOMCAT, show substantially slower vertical mixing in the troposphere compared to other models (LMDZ, TM5, EMAC, and ACTM). However, while the TOMCAT model, as used here, has slow transport between the hemispheres and between land and ocean, the NIES model shows efficient horizontal mixing and a smaller latitudinal gradient in SF<sub>6</sub> compared to the other models and observations. We find consistent differences between models concerning vertical mixing of the troposphere, expressed as AoA differences and modeled <sup>222</sup>Rn gradients between 950 and 500 hPa. All models agree, however, on an interesting asymmetry in inter-hemispheric mixing, with faster transport from the Northern hemisphere surface to the Southern hemisphere than vice versa. This is attributed to a rectifier caused by a stronger seasonal cycle in boundary layer venting over Northern hemispheric landmasses, and possibly to a related asymmetric position of the inter-tropical convergence zone. The calculated AoA in the upper stratosphere varies considerably among the models (4 –7 years). Finally, we find that the inter-model differences are generally larger than differences in AoA that result from using the same model with a different resolution or convective parameterisation. Taken together, the AoA model inter-comparison provides a useful addition to traditional approaches to evaluate transport time scales. Results highlight that inter-model differences



associated with resolved transport (advection) and parameterised transport (convection, boundary layer mixing) are still large, and require further analysis. For this purpose, all model output and analysis software is available.

## 1 Introduction

The composition of the atmosphere is determined by exchange processes at the Earth's surface, transport processes within the atmosphere, and chemical and physical conversion processes. For example, the atmospheric mixing ratio distribution of CH<sub>4</sub> is driven by natural and anthropogenic emissions at the Earth's surface, atmospheric transport, and removal that is primarily driven by the atmospheric oxidants OH, Cl, and O(<sup>1</sup>D) (Myhre et al., 2013). The estimated atmospheric lifetime of CH<sub>4</sub> is approximately 9 years (Prather et al., 2012) mainly due to oxidation within the troposphere. Although some CH<sub>4</sub> destruction occurs in the stratosphere, as witnessed by the decaying mixing ratios with altitude, the slow atmospheric transport from the troposphere to the stratosphere limits its impact on the atmospheric lifetime of CH<sub>4</sub>. Other atmospheric constituents have widely different budgets. SF<sub>6</sub>, for instance, has purely anthropogenic sources, mostly in the Northern Hemisphere (NH), and is only broken down in the upper stratosphere, resulting in a very long atmospheric lifetime of more than 1000 years (Kovács et al., 2017). In contrast, <sup>222</sup>Rn is of purely natural origin. It emanates from land surfaces and quickly decays radioactively with a half life of 3.8 days.

To better understand the changes in the atmosphere, General Circulation Models (GCMs) or Chemistry Transport Models (CTMs) are used to simulate its composition. While CTMs use archived meteorological fields to calculate transport, GCMs calculate their own transport. To track the observed state of the atmosphere, the GCMs can be nudged towards meteorological re-analysis data (Law et al., 2008). Comparing results of these models to atmospheric observations allows assessment of the model performance. Depending on the atmospheric compound, various aspects of atmospheric transport can be investigated. Jacob et al. (1997) used <sup>222</sup>Rn and other short-lived tracers to evaluate the convective and synoptic scale transport in CTMs. Since the early 1990s, the atmospheric tracer transport model intercomparison project (TransCom) has carried out studies to quantify and diagnose the uncertainty in inversion calculations of the global carbon budget that result from errors in simulated atmospheric transport. Initially, TransCom focused on non-reactive tropospheric species such as SF<sub>6</sub> (Denning et al., 1999) and CO<sub>2</sub> (Law et al., 1996, 2008). A more recent TransCom model inter-comparison (Patra et al., 2011) focused on the ability of models to properly represent atmospheric transport of CH<sub>4</sub>, focussing on vertical gradients, its average long-term trends, seasonal cycles, inter-annual variations (IAVs) and inter-hemispheric (IH) gradients. The study concluded that models that show faster IH exchange for SF<sub>6</sub>, also exhibit smaller IH gradients in CH<sub>4</sub>. The estimated IH exchange time, calculated based on a time-invariant SF<sub>6</sub> emission rate, remained relatively constant over the period of the analysis (1996–2007). Interestingly, a recent study highlighted the importance of IH transport variations in explaining the CO<sub>2</sub> mixing ratio difference between Mauna Loa in the Northern Hemisphere (NH) and Cape Grim, Tasmania, in the Southern Hemisphere (SH) (Francey and Frederiksen, 2016). Specifically, the 0.8 ppm step-like increase of this difference between 2009 and 2010 was attributed to the opening and closing of the upper-tropospheric equatorial westerly duct, with an open-duct pattern from July 2008 to June 2009 (fast IH exchange), followed by closed-duct conditions between from July 2009 to June 2010 (slow IH exchange). Confirming



this mechanism, Pandey et al. (2017) found again faster IH transport of CH<sub>4</sub> during the strong La Niña in 2011 using the TM5 CTM. The time-scale of IH transport is an important parameter for atmospheric inversion studies. In these studies atmospheric observations are used to infer surface flux magnitude and distribution. For a long-lived greenhouse gas like CH<sub>4</sub> fast IH transport implies that a larger fraction of the emissions will be attributed to the NH (Patra et al., 2014, 2011).

5 The efficiency of models to mix the planetary boundary and to vent emissions to the overlying free atmosphere has been studied using SF<sub>6</sub> and generally revealed too slow mixing over mid-latitude continents in the TM5 (Peters et al., 2004) and MOZART models (Gloor et al., 2007). Together with the convective parameterisation, this determines the rate by which emissions (e.g. SF<sub>6</sub> and <sup>222</sup>Rn) are mixed vertically and inter-hemispherically (Locatelli et al., 2015a).

Another transport time scale relevant for atmospheric composition studies is troposphere-stratosphere exchange, driven by  
10 the Brewer-Dobson circulation (Butchart, 2014). Depending on the greenhouse gas scenario, global climate model projections predict an acceleration of this global mass circulation of tropospheric air through the stratosphere. Stratospheric age of air and its temporal trend have been determined from SF<sub>6</sub> measurements from the MIPAS satellite (Stiller et al., 2012). Using a suite of stratospheric observations, Fu et al. (2015) quantified the acceleration of the Brewer-Dobson circulation as 2.1% per decade for 1980–2009. Other modelling and experimental studies revealed that the atmospheric composition of the tropopause layer  
15 strongly depends on the mixing processes that occur on a wide range of spatiotemporal scales (Hoor et al., 2010; Berthet et al., 2007; Prather et al., 2011)

Transport of trace gases in CTMs and GCMs is determined by several factors, for which various choices are possible. First, winds and the choice of advection scheme drive the large-scale dispersion and IH transport of tracers. Second, parameterised sub-grid-scale processes such as boundary layer mixing and convection determine vertical gradients and the rate of IH transport  
20 (e.g., Locatelli et al., 2015a). Finally, other differences may be caused by the horizontal and vertical grid of the model, and other issues related to spatial and temporal integration. For instance, by just doubling the vertical resolution of their model, Locatelli et al. (2015a) largely improved the representation of SF<sub>6</sub> transport from the troposphere to the stratosphere. Likewise, Bândă et al. (2015) reported large differences in the stratospheric dispersion of the 1991 Pinatubo plume when increasing the vertical resolution in TM5 from 34 to 60 levels.

25 To investigate the impact of these choices in the participating models, this paper will present the first results of the TransCom age-of-air (AoA) inter-comparison study. The concept age-of-air originates from stratospheric studies (Hall and Plumb, 1994; Neu and Plumb, 1999; Hall et al., 1999). In brief, the age spectrum in the stratosphere  $G(x, t|t_0)$  is calculated as a type of Green's function that propagates a tropospheric mixing ratio boundary condition into the stratosphere. Given a location  $x$  in the stratosphere,  $G\delta t$  represents the fraction of air at  $x$  that was lost in the troposphere in the time interval between  $t - t_0$  and  
30  $t - t_0 + \delta t$  (Hall et al., 1999). In practical model applications focussing on stratospheric age spectra,  $G(x, t|t_0)$  is calculated as the response of a time-dependent boundary condition  $\delta(t - t_0)$  specified in a forcing volume in the troposphere. More recently, this concept has also been applied to tropospheric studies (Holzer and Hall, 2000; Waugh et al., 2013; Holzer and Waugh, 2015). In this AoA inter-comparison, we will only analyse the mean age of air. This property can be easily extracted from model simulations of a passive tracer with linearly growing boundary conditions in a specified atmospheric volume. We will

**Table 1.** The five AoA tracers and their forcing volume.

AoA Tracer	Forcing volume
Surface	Surface < 100 m
NHsurface	NH surface <100 m
SHsurface	SH surface <100 m
Land	Land < 100 m
Ocean	Ocean < 100 m

outline the inter-comparison protocol in section 2. The resulting model output focussing on the simulation period 2000–2011 is compared in section 3, and discussed in section 4. Finally, the main conclusions are summarised in section 5.

## 2 Method

### 2.1 AoA protocol

5 In order to compare transport time scales of a suite of CTMs and GCMs, a protocol was developed that allows a straightforward implementation in existing atmospheric models. We defined five AoA tracers (Table 1) for which linearly growing boundary conditions are applied. These AoA tracers are chosen to study IH transport, transport to the stratosphere, and air mass transport between land and ocean. The mixing ratios of these tracers in the models are initialised to zero on January 1, 1988 and simulations are run to the end of 2014.

10 According to the protocol, the mixing ratio in the forcing volume is set every time step to a value  $B = f \times t$ , with  $t$  the elapsed time (in s) since January 1988, and  $f$  a forcing constant of  $1 \times 10^{-15} \text{ mol mol}^{-1} \text{ s}^{-1}$ . As a result, the mixing ratio in the forcing volume will have a value of  $852.0768 \text{ nmol mol}^{-1}$  at the end of the simulation, and lower values elsewhere. In theory, numerical issues with advection might lead to small wiggles in the vicinity of strong mixing ratio gradients. This may result in small unphysical negative mixing ratios that cannot be handled by some model transport schemes. To remedy this, models may be initialised with a uniform  $100 \text{ nmol mol}^{-1}$  initial condition, a value that is subtracted before further analysis.

Modellers are requested to calculate the exact fraction of the grid box within the forcing volume. This calculation involves the land mask, the fraction of the grid box on either the NH and SH, and the geopotential height. The protocol provides example code to help with the implementation. Importantly, the mixing ratio of the grid boxes within the forcing volume was set according to:

$$20 \quad X_{new} = f_{set} \times B + (1 - f_{set}) \times X_{old}, \quad (1)$$

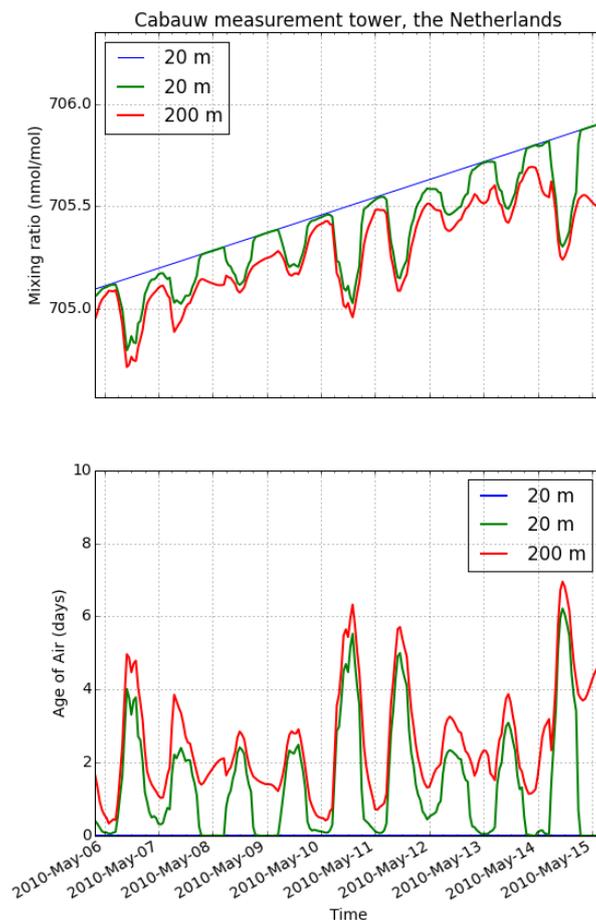
**Table 2.** Additional tracers in the AoA intercomparison.

Tracer	Remarks
$^{222}\text{Rn}$	Radon tracer, similar to Patra et al. (2011)
$^{222}\text{RnE}$	As $^{222}\text{Rn}$ , but with specific monthly emissions over Europe in 2006-2010 (Karstens et al., 2015)
$\text{SF}_6$	Similar to Patra et al. (2011), but with updated yearly emissions (Table 3)
E90	Tracer with surface emissions and atmospheric lifetime of 90 days (Prather et al., 2011)

where  $f_{set}$  is the fraction of the grid box within the forcing volume, and  $X_{old}$  the mixing ratio before the forcing procedure. Note that in locations where  $f_{set} = 0$ , nothing needs to be changed, and the mixing ratio changes are purely driven by transport from the forcing volume. To diagnose AoA, the simulated mixing ratios  $X$  in the atmosphere can be converted into AoA by  $L = t - \frac{X}{f}$ , with  $L$  the AoA in seconds and  $t$  the elapsed time of the simulation.

5 Figure 1 shows the simulated mixing ratios and applied forcing for the tracer "Surface" at the location of the Cabauw tower from May 6 to May 15, 2010, together with the derived AoA. We use here output from the TM5 model on  $1 \times 1$  degree resolution (see Table 4). At 20 m above the surface during night-time AoA is generally close to zero, due to the fact that the volume is forced in the lowest 100 m, and vertical mixing is limited in a stable nocturnal boundary layer. During daytime, however, older air from aloft is mixed in, and the AoA increases depending on the depth of the mixing layer and the strength  
 10 of the vertical mixing. At 200 m, outside the forcing volume, air is generally older. In some nights, e.g. from May 8 to May 9, the surface gets decoupled from the air masses aloft, signalling a stable boundary layer. Note that the depth of the lowest model layer is approximately 20 m, which implies that this layer is entirely within the forcing volume (the applied forcing is indicated by the blue line). However, TM5 still calculates a non-zero AoA in the lowest model layer, because the mixing ratios are sampled after vertical transport, which mixes in older air (Krol et al., 2005). We do not provide recommendations on the  
 15 sampling strategy. For instance, models that sample right after forcing, simulate a linear mixing ratio increase at 20 m (blue line in the upper panel of Figure 1) and a constant and zero AoA at 20 m (blue line in the lower panel of Figure 1). The current paper will focus on large-scale transport timescales and we found no major influence of this sampling strategy on the results.

To complement the AoA tracers, modellers were also requested to simulate the tracers listed in Table 2. Simulations of  $^{222}\text{Rn}$  are intended to study vertical and synoptic scale transport (Jacob et al., 1997);  $\text{SF}_6$  is used to diagnose IH transport,  
 20 stratosphere–troposphere exchange, and stratospheric AoA; E90 is used to diagnose the "chemical" tropopause and will be compared to Prather et al. (2011). Note that we also included a  $^{222}\text{Rn}$  simulation with monthly varying emissions over Europe during 2006–2010, based on the high resolution emissions maps presented in Karstens et al. (2015). The current paper will, however, not analyse these simulations.



**Figure 1.** Upper panel: Mixing ratios of an AoA tracer simulated by the TM5 model (TM5\_1x1) forced at the surface along the Cabauw measurement tower in the Netherlands during May 2010. The blue line represents the applied forcing. Lower panel: Similar, but transferred into Age of Air.

Homogeneous surface emissions (in  $\text{kg m}^{-2} \text{s}^{-1}$ ) of E90 ( $E_{E90}$ ) are calculated such that the mean steady-state atmospheric mixing ratio of E90 will approach  $100 \text{ nmol mol}^{-1}$ :

$$E_{E90} = \frac{M_{atm} \times 100 \times 10^{-9}}{\tau_{E90} \times 4\pi r^2} \quad (2)$$

with  $M_{atm}$  the atmospheric mass (kg),  $r$  the radius of the Earth (m), and  $\tau_{E90}$  the e-folding lifetime (90 days in units s) of E90.

Emissions of  $\text{SF}_6$  based on EDGAR4.0 with corrections suggested by Levin et al. (2010) are given in Table 3. The emission distribution is similar to Patra et al. (2011).

**Table 3.** Yearly emissions of SF<sub>6</sub> for the period 1988-2015.

Year	Source (mmol s <sup>-1</sup> )	Year	Source (mmol s <sup>-1</sup> )
1988	934	2002	1223
1989	938	2003	1258
1990	1036	2004	1268
1991	1116	2005	1299
1992	1210	2006	1366
1993	1303	2007	1475
1994	1381	2008	1555
1995	1392	2009	1577
1996	1312	2010	1599
1997	1208	2011	1642
1998	1162	2012	1685
1999	1177	2013	1729
2000	1201	2014	1772
2001	1197	2015	1816

NetCDF-formatted input files were provided with emissions on 1 × 1 degree resolution and initial SF<sub>6</sub> mixing ratios for January 1, 1988. In terms of output, modellers were requested to provide monthly mean mixing ratios and hourly mixing ratio time series at 247 atmospheric measurement stations. Furthermore, hourly atmospheric profiles of mixing ratios and meteorological variables (u-wind, v-wind, surface pressure, boundary layer height, geopotential height) were requested at 119 locations. In this first analysis we will concentrate on the output of monthly mean mixing ratios that have been provided by six CTMs, some of them running in different configurations. Table 4 lists the participating models along with their horizontal and vertical resolution. Three models (LMDZ, EMAC, ACTM) run in "on-line" mode, meaning that meteorology is calculated by the physics module of the model, and nudged towards data of a reanalysis (ECMWF Era interim, or JMA). Three other models (TM5, NIES, TOMCAT) run in "off-line" mode, and directly read in meteorological driver data from a reanalysis. TM5\_EC-Earth reads in meteorological data from the EC-Earth model (Hazeleger et al., 2010) that is nudged to Era Interim (see Section 2.3)

In the next subsections, specific information on the participating models and the measurements is given.

## 2.2 LMDZ

LMDz, developed at Laboratoire de Météorologie Dynamique with "z" standing for zoom capacity, is the global general circulation model of the IPSL Earth system model (Hourdin et al., 2006, 2012). Here, we include two offline versions of LMDz – LMDZ3 and LMDZ5A –, both with a horizontal resolution of 1.875° (latitude) × 3.75° (longitude) and a vertical resolution

**Table 4.** Short-hand notation of the models participating in this study, along with model information.

Model submission	Base model	longitude × latitude	Vertical levels	Meteorological Driver Data
LMDZ3	LMDZ	$3.75^\circ \times 1.875^\circ$	39 hybrid $\sigma$ -pressure	Nudged to Era interim
LMDZ5A	LMDZ	$3.75^\circ \times 1.875^\circ$	39 hybrid $\sigma$ -pressure	Nudged to Era interim
TM5_3x2	TM5	$3^\circ \times 2^\circ$	60 hybrid $\sigma$ -pressure	Era interim
TM5_1x1	TM5	$1^\circ \times 1^\circ$	60 hybrid $\sigma$ -pressure	Era interim
TM5_EC-Earth	TM5	$3^\circ \times 2^\circ$	34 hybrid $\sigma$ -pressure	EC-Earth, nudged to Era interim
EMAC_T63	EMAC	$\approx 1.875^\circ \times 1.875^\circ$	90 hybrid $\sigma$ -pressure	Nudged to Era interim
EMAC_T106	EMAC	$\approx 1.125^\circ \times 1.125^\circ$	90 hybrid $\sigma$ -pressure	Nudged to Era interim
ACTM	ACTM	$\approx 2.81^\circ \times 2.81^\circ$	67* $\sigma$ up to 90 km	Nudged to JMA Reanalysis
NIES	NIES	$2.5^\circ \times 2.5^\circ$	32 hybrid $\sigma - \theta$ up to 5 hPa	JRA-25/JCDAS
TOMCAT	TOMCAT	$\approx 2.81^\circ \times 2.81^\circ$	60 hybrid $\sigma$ -pressure	Era interim

\*Only the lower 50 layers have been submitted

of 39 hybrid sigma-pressure levels, commonly chosen for global inverse studies using LMDz (Chevallier, 2015; Locatelli et al., 2015b; Yin et al., 2017). These two versions are different in terms of physical parameterisation of deep convection and boundary layer mixing. LMDZ3 uses the deep convection scheme of Tiedtke (1989) and the boundary layer mixing scheme of Louis (1979). LMDZ5A is an updated version that uses the deep convection scheme of Emanuel (1991) and the boundary layer mixing parameterisation from Louis (1979), further adjusted according to Deardorff (1966). More details regarding the configurations of the physics and comparison to other versions are described in Locatelli et al. (2015a). Sea surface temperature and sea ice coverage from the ERA-Interim climate reanalysis (Dee et al., 2011) produced by the European Centre for Medium-Range Weather Forecasts (ECMWF) are used as boundary conditions. Horizontal winds are nudged towards the ERA-Interim wind fields with a relaxation time of 3 hours.

### 10 2.3 TM5

The TM5 model (Krol et al., 2005) has different application areas with a common core. TM5 allows for a flexible grid definition with two-way nested zoom regions. This version is mainly used in inverse modelling applications, focussing on CO<sub>2</sub> (Peters et al., 2010), CH<sub>4</sub> (Houweling et al., 2014; Bergamaschi et al., 2013), and CO (Krol et al., 2013). The chemistry version of TM5 (Huijnen et al., 2010) was recently adopted for massive parallel computing (Williams et al., 2017). The AoA simulations were conducted in this so-called TM5-MP version.

The TM5 model is used in three versions that differ in horizontal resolution and in the way meteorological data is used. The version named TM5\_3x2 simulated the AoA experiment at a resolution of  $3^\circ \times 2^\circ$  (longitude × latitude). In the version TM5\_1x1, a horizontal resolution of  $1^\circ \times 1^\circ$  was used. TM5\_1x1 and TM5\_3x2 refer to simulations with the off-line TM5-MP version that reads in the meteorological fields from files. Like earlier model versions, TM5\_1x1 and TM5\_3x2 are driven



by the ERA-Interim meteorological re-analysis (Dee et al., 2011) and updated every 3 h, with time-interpolation during time integration. For this inter-comparison we include all 60 vertical levels provided by the ECMWF ERA-Interim reanalysis. Convective mass fluxes are derived from entrainment and detrainment rates from the ERA-Interim dataset. This replaces the convective parameterisation used in the previous TransCom inter-comparison (Patra et al., 2011), which was based on Tiedtke (1989). Recent analysis (Tsuruta et al., 2016) shows that the mass fluxes produced with the ERA-interim data set (Dee et al., 2011) lead to faster inter-hemispheric transport compared to the old model version using the Tiedtke (1989) scheme that was used in the earlier TransCom study (Patra et al., 2011).

The vertical diffusion in the free troposphere is calculated according to Louis (1979), and in the boundary layer by the approach of Holtslag and Boville (1993). Diurnal variability in the boundary layer height is determined using the parameterisation of Vogelesang and Holtslag (1996). Advective fluxes are calculated using the slopes scheme (Russel and Lerner, 1981), with a refinement in time step whenever the Courant–Friedrichs–Lewy (CFL) criterion is violated.

TM5\_EC-Earth is the atmospheric transport model of the Earth System Model EC-Earth version 3.2.2 (Version 2.3 is described in Hazeleger et al. (2010, 2012)) The main differences between the two versions are an increased resolution (from T159 with 62 vertical layers in version 2.3 to T255 with 91 vertical layers in version 3.2.2) and updated versions of the separate model components. Only the atmospheric components are used in this study. The dynamical core of EC-Earth is based on the Integrated Forecasting System (IFS), version cy36r4 (Molteni et al., 2011). In contrast to the offline versions of TM5, the dynamical properties of the atmosphere are simulated by EC-Earth in TM5\_EC-Earth. EC-Earth was nudged towards ERA-Interim data (Dee et al., 2011) for temperature, vorticity, divergence, and the logarithm of the surface pressure. The nudging used a relaxation time of 6 hours, allowing for continuous atmospheric dynamics while following the ERA-Interim data. Every 6 hours, the meteorological data is transferred via de OASIS3 coupler (Valcke, 2013) to TM5-MP, which uses this data to calculate the actual atmospheric transport as described above. Apart from the meteorological data, the model code of TM5-EC-Earth is similar to TM5\_3x2, but with a reduced number of vertical levels (34, i.e. a subset of the 91 levels used in IFS, instead of 60).

## 2.4 EMAC

The EMAC (ECHAM/MESSy Atmospheric Chemistry) model employed in this study, combines an updated version 2.50 of the MESSy (Modular Earth Submodel System) framework (Jöckel et al., 2005, 2010) with version 5.3.02 of the ECHAM5 (European Centre Hamburg) general circulation model (Roeckner et al., 2006). Our version of the EMAC model, first described by Jöckel et al. (2006), incorporates a recent update to convective transport of tracers (Ouwensloot et al., 2015) and is further improved upon to facilitate nudging of tracers according to Table 1. These modifications are included in version 2.52 of MESSy.

Dynamical properties are simulated by EMAC itself. The model dynamics has been weakly nudged in the spectral space, nudging temperature, vorticity, divergence and surface pressure (Jeuken et al., 1996). Different nudging coefficients are used for different vertical levels, with no nudging in the boundary layer and above ~10 hPa, and with maximum nudging in the free troposphere. Convective mass fluxes are diagnosed in the CONVECT submodel (Tost et al., 2006), while the resulting transport is calculated by the CVTRANS model (Tost et al., 2010; Ouwensloot et al., 2015). The simulations with EMAC make use of



90 vertical hybrid sigma pressure levels. Data is available for two horizontal resolutions: T63 ( $192 \times 96$  grid) with a fixed time step of 6 min, and T106 ( $320 \times 160$  grid) with a fixed time step of 4 min.

## 2.5 ACTM

The CCSR/NIES/FRCGC (Center for Climate System Research/National Institute for Environmental Studies/Frontier Research Center for Global Change) atmospheric general circulation model (AGCM)-based chemistry-transport model (ACTM) is developed for simulations of long-lived gases in the atmosphere (Numaguti et al., 1997; Patra et al., 2009a, 2014). The ACTM simulations are performed at a horizontal resolution of T42 spectral truncation ( $\sim 2.8 \times 2.8$  deg) with 67 sigma levels in the vertical and model top at  $\sim 90$  km. The horizontal winds and temperature of ACTM are nudged with Japan Meteorological Agency Reanalysis (JRA) data products (Onogi et al., 2007). The nudging forces the AGCM-derived meteorology towards the reanalysed horizontal winds (U and V components) and temperature (T) with relaxation times of 2 and 5 days, respectively, except for the top and bottom model layers.

The heat and moisture exchange fluxes at the Earth's surface are calculated using inter-annually varying monthly-mean sea ice and sea-surface temperature from the Hadley Centre observational data products (Rayner et al., 2003). Tracer advection is performed using a 4th order flux-form advection scheme comprising of monotonic piecewise parabolic method (Colella and Woodward, 1984) and a flux-form semi-Lagrangian scheme (Lin and Rood, 1996).

Sub-grid-scale vertical fluxes are approximated using a non-local closure scheme based on Holtslag and Boville (1993) and the level 2 scheme of Mellor and Yamada (1974). The cumulus parameterisation scheme is simplified from Arakawa and Schubert (1974) and used for calculating the updraft and downdraft of tracers by cumulus convection.

## 2.6 NIES

The National Institute for Environmental Studies (NIES) Eulerian three-dimensional offline transport model is driven by the Japanese Meteorological Agency Climate Data Assimilation System (JCDAS) datasets (Onogi et al., 2007). It employs a reduced horizontal latitude–longitude grid with a spatial resolution of  $2.5^\circ \times 2.5^\circ$  near the equator (Maksyutov and Inoue, 1999) and a flexible hybrid sigma–isentropic ( $\sigma$ – $\theta$ ) vertical coordinate, which includes 32 levels from the surface up to 5 hPa (Belikov et al., 2013b). The vertical transport for all levels above tropopause and higher than a potential temperature level of 295 K follows a climatological adiabatic heating rate. The parameterisation of turbulent diffusivity separates transport processes in the planetary boundary layer (provided by the ECMWF ERA-Interim reanalysis) from the free troposphere following the approach by Hack et al. (1993). Vertical mass fluxes due to cumulus convection are based on the convective precipitation rate provided by the reanalysis dataset (Austin and Houze Jr, 1973; Belikov et al., 2013a). A modified Kuo-type parameterization scheme (Grell et al., 1994) is used to set cloud top and cloud bottom height. Transport by convective updrafts and downdrafts includes entrainment and detrainment processes as described by Tiedtke (1989).



## 2.7 TOMCAT

TOMCAT/SLIMCAT is a global 3-D off-line chemical transport model (Chipperfield, 2006). It is used to study a range of chemistry-aerosol-transport issues in the troposphere (e.g., Monks et al., 2017) and stratosphere (e.g., Chipperfield et al., 2015). The model is usually forced by ECMWF meteorological (re)analyses, although GCM output can also be used. When using ECMWF fields, as in the experiments described here, the model reads in the 6-hourly fields of temperature, humidity, vorticity, divergence and surface pressure. The resolved vertical motion is calculated online from the vorticity. The model has different options for the parameterisations of sub-grid-scale tracer transport by convection (Stockwell and Chipperfield, 1999; Feng et al., 2011) and boundary layer mixing (Louis, 1979; Holtslag and Boville, 1993). Tracer advection is performed using the conservation of second-order moments scheme of Prather et al. (1987). For the experiments the model was run at horizontal resolution of  $2.8^\circ \times 2.8^\circ$  with 60 hybrid  $\sigma$ -pressure levels from the surface to ~60 km, which follow the vertical levels from the meteorological fields from ERA-Interim reanalyses (Dee et al., 2011). The model was forced by ECMWF ERA-Interim reanalyses. Convective mass fluxes were diagnosed online using a version of the Tiedtke scheme (Stockwell and Chipperfield, 1999) and mixing in the boundary layer is based on the local scheme of Louis (1979). Previous work with the model has shown that these schemes tend to underestimate the mixing out of the boundary layer and convective transport to the upper troposphere (e.g. Feng et al., 2011) but these were the options available for the multi-decadal runs analysed here.

## 2.8 Measurement data

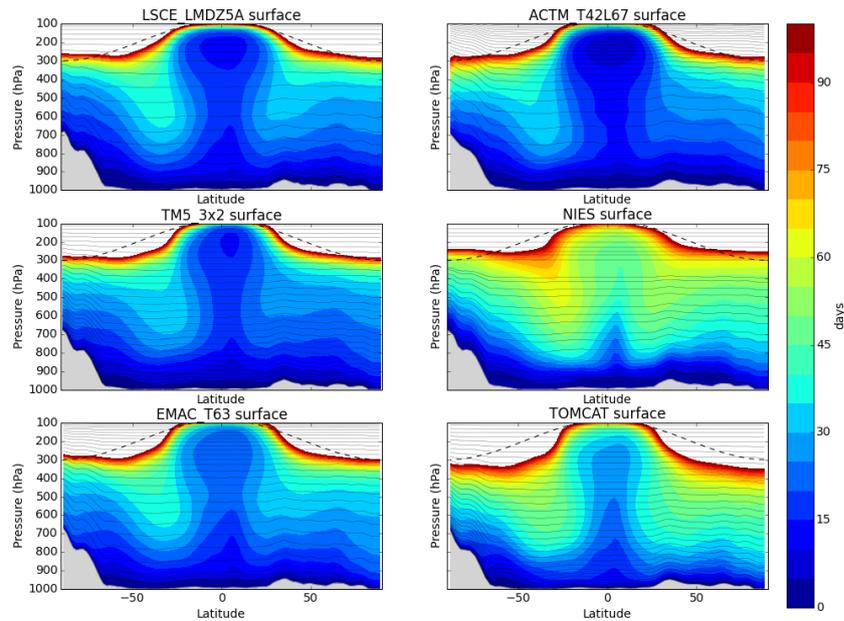
We will compare model results to latitudinal SF<sub>6</sub> gradients measured by the National Oceanic & Atmospheric Administration Earth System Research Laboratory (NOAA/ESRL) (Hall et al., 2011). We use the combined data set constructed from flask data measured by the Halocarbons and other Atmospheric Trace Species (HATS) group and hourly CATS data (downloaded from <https://www.esrl.noaa.gov/gmd/hats/combined/SF6.html>), and include only stations with a full measurement record in the period 2000–2011. Subsequently, the difference with respect to the South Pole is calculated as an 11-year time series with monthly time resolution. The mean and standard deviations of these time series are added to the modeled latitudinal gradients below.

In Appendix A we further compare the simulated SF<sub>6</sub> latitudinal and vertical gradients to measurements made during the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observation (HIPPO) campaigns (Wofsy, 2011), similar to comparisons presented in Patra et al. (2014).

## 3 Results

### 3.1 Tropospheric AoA

First, we focus on zonal and multi-year averages to investigate differences among the models in IH and vertical transport in the troposphere. Monthly mean mixing ratios (and pressure fields) of the participating models have been averaged zonally and

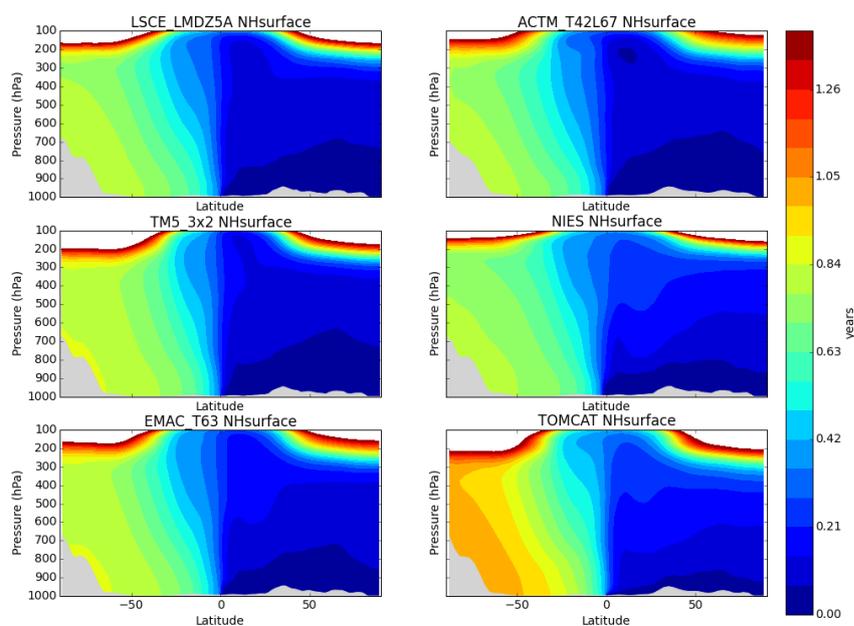


**Figure 2.** Zonally averaged AoA (days) in the troposphere for tracer "Surface". The results have been averaged over the period 2000–2010 (11 years). The light grey areas correspond to the zonal mean orography in the models. The thin black lines denote the mid-pressure levels of the models. For reference, the dotted black line in all panels denotes the climatological tropopause (Lawrence et al., 2001). The white areas correspond to areas in which the air is older than 100 days.

over time, and converted to AoA. Contour plots of these latitude–pressure arrays are presented in Figures 2, 3, and 4 for the AoA tracers Surface, NHsurface, and SHsurface, respectively (see Table 1).

The AoA in the troposphere derived from tracer "Surface" is generally younger than 100 days (Figure 2). The 100 day contour sharply marks the transition to the stratosphere at around 300 hPa at the poles. In the tropics this transition is located at a pressure lower than 100 hPa. This generally agrees with the definition of the climatological tropopause given by Lawrence et al. (2001). Deep convective transport drives the vertical transport in the tropics, and this deep convective region is bounded by tongues of older air connected to the stratosphere, signalling stratosphere-troposphere exchange. Although all models agree on this general pattern, clear differences are also present. Deep convective mixing in the tropics is strongest in ACTM, while TOMCAT and NIES generally show slower vertical mixing and larger AoA gradients between the surface and the upper troposphere.

Tracers "NHsurface" (Figure 3) and "SHsurface" (Figure 4) are used to diagnose IH transport. Generally, the air at high latitudes has an age between 0.6 and 1.2 years in the hemisphere opposite to the forcing volume. Here, all models agree on an interesting asymmetry: AoA derived from "SHsurface" around the North Pole is older than AoA derived from "NHsurface"

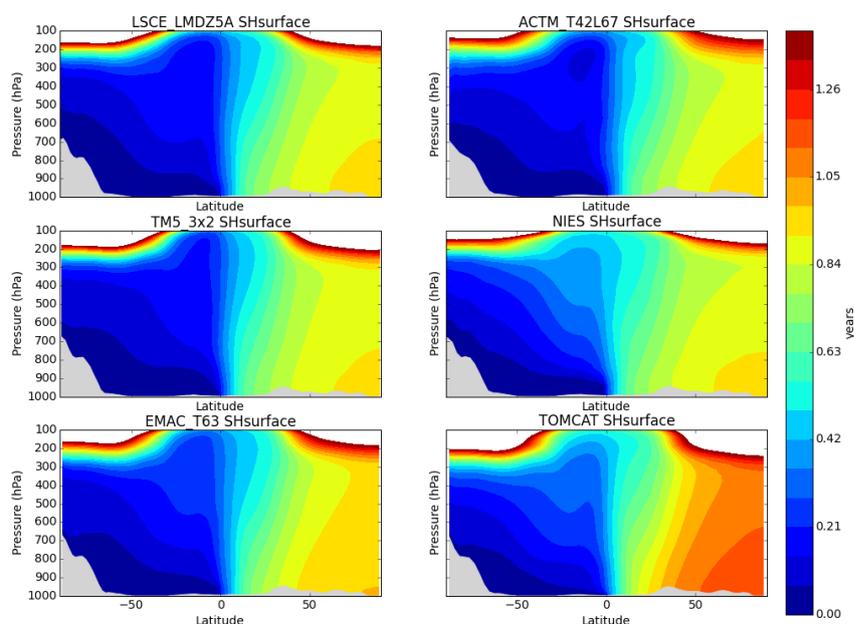


**Figure 3.** Zonally averaged AoA (years) in the troposphere for tracer "NHsurface". The results have been averaged over the period 2000–2010 (11 years). The light grey areas correspond to the zonal mean orography in the models. The white areas correspond to regions in which the air is older than 1.4 years.

around the South Pole. TOMCAT, diagnosed with slow vertical mixing, has old air around at high latitudes, while NIES (also slow vertical mixing) has high-latitude ages more in line with other models, indicating fast horizontal transport near the surface. Figures 3 and 4 illustrate the fact that exchange of air between the hemispheres proceeds faster at higher altitudes (200–500 hPa), and consequently steep AoA gradients are observed close to the surface in the tropics.

### 5 3.2 AoA derived from "Land" and "Ocean" tracers

Figures 5 and 6 show AoA derived from tracers "Land" and "Ocean" evaluated in the lowest model level. Oldest air related to "last contact with land" is found over the Southern Oceans, with ages older than 100 days. Models generally agree on the fact that old air is found in the stationary high-pressure areas over the ocean that are related to the Hadley and Walker circulations. The TOMCAT model, in the configuration used here, has systematically larger AoA at the surface. Dominant land-ocean transport patterns are easily discerned in Figure 5. Young air over oceans is found south-east of South America, north-west of Australia, and towards the north-east of North-America and Asia, associated with the position of the upper-air jet-stream. The oldest air in the southern East Pacific for tracer "Land" indicates that this region is most isolated from the emission from the



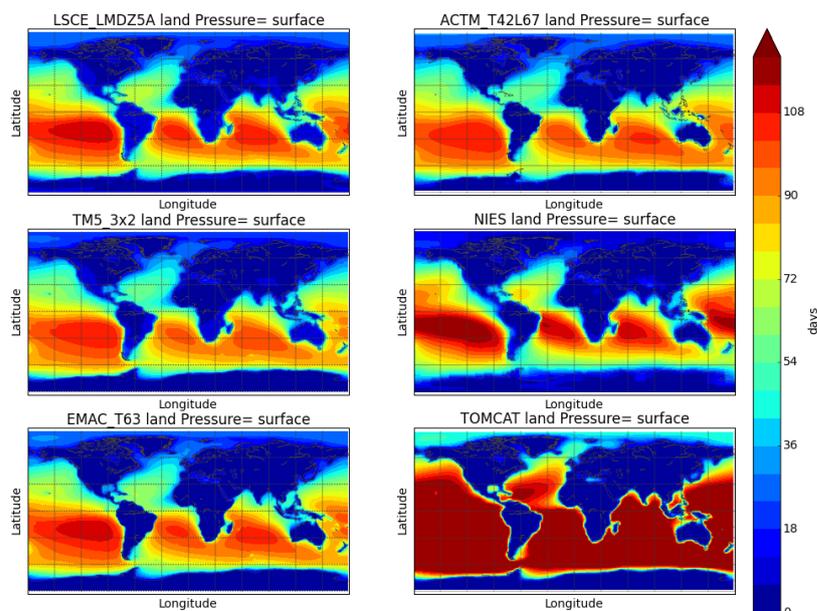
**Figure 4.** Zonally averaged AoA (years) in the troposphere for tracer "SHsurface". The results have been averaged over the period 2000–2010 (11 years). The light grey areas correspond to the zonal mean orography in the models. The white areas correspond to regions in which the air is older than 1.4 years.

NH. Indeed, the lowest  $\text{CH}_4$  concentrations on the Earth's surface are found at NOAA site Eastern Island (EIC) (Patra et al., 2009b), which was attributed to "old" air in combination with strong removal of  $\text{CH}_4$  by OH at tropical latitudes.

The AoA derived from tracer "Ocean" (Figure 6) generally shows ages less than 60 days over land, with the older ages logically located in areas deep inland. Although these patterns are similar in all models, the spread is considerable, with TOMCAT again showing the oldest air, and TM5 showing the youngest air over land surfaces. These differences are likely related to the boundary layer parameterisations in the models, and in the case of TOMCAT the use of the simple local Louis (1979) scheme. The AoA of tracer "Ocean" over land is strongly determined by dominant circulation patterns such as the monsoon, trade winds, and jet-streams.

### 3.3 Stratospheric AoA

We use tracer "Surface" to compare the simulated stratospheric AoA. Here it should be noted that the TOMCAT and NIES AoA is already systematically older at the tropopause (see Figure 2). In Figure 7 the stratospheric AoA is plotted for all models from 100 hPa to the top of the atmosphere, averaged over the period 2000–2010 (11 years). As expected, oldest AoA is found at the high-altitude poles. Considerable model spread is found with the oldest air (up to 7 years) in LMDZ5A and

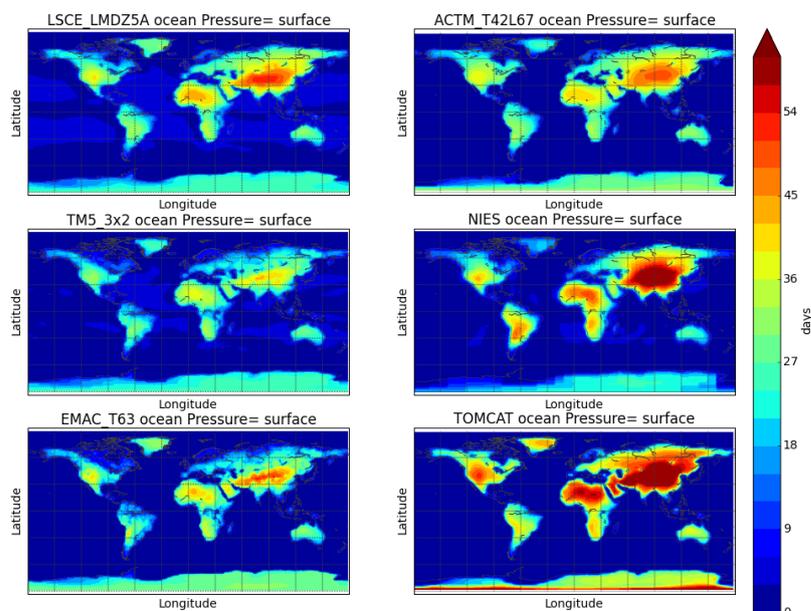


**Figure 5.** Latitude-longitude plot of the AoA (days) derived from tracer "Land" evaluated in the lowest model layer. The results have been averaged over the period 2000-2010 (11 years). Ages older than 120 days appear in dark red.

the youngest air in ACTM (< 5 years). The transport in EMAC, TOMCAT, LMDZ, and TM5 is driven by – or nudged to – ERA-interim meteorology (see Table 4), and one would expect similar stratospheric AoA in these models. However, AoA in the stratosphere is not only determined by the driving meteorological data, but also by the treatment of advection (specifically vertical transport), nudging parameters, and the number of vertical layers in the model. According to a stratospheric AoA study by Diallo et al. (2012) the use of instantaneous wind fields at 3 or 6-hourly time intervals in CTMs may under-sample fast vertical variations during these time intervals. For GCMs that are nudged to reanalysis meteorological data, gravity wave noise may influence the stratospheric AoA. Finally, the vertical coordinate system may influence numerical transport effects (Diallo et al., 2012). Further analysis on the stratospheric AoA in this model ensemble is left for future exploration.

### 3.4 Interhemispheric transport

10 In this section, we compare the inter-hemispheric transport times using tracers "NHsurface" and "SHsurface" to the simulated latitudinal gradients of SF<sub>6</sub>. To this end, we show in Figure 8 (upper panel) the simulated zonal average latitudinal SF<sub>6</sub> gradient at the surface. All gradients have been scaled relative to the South Pole SF<sub>6</sub> mixing ratio. Also included in the figure are the latitudinal gradients measured by NOAA/ESRL (Hall et al., 2011). Since the selected model results are averages over all longitudes, including the land masses with high emissions, modeled zonal averages exceed the observations at NH mid-

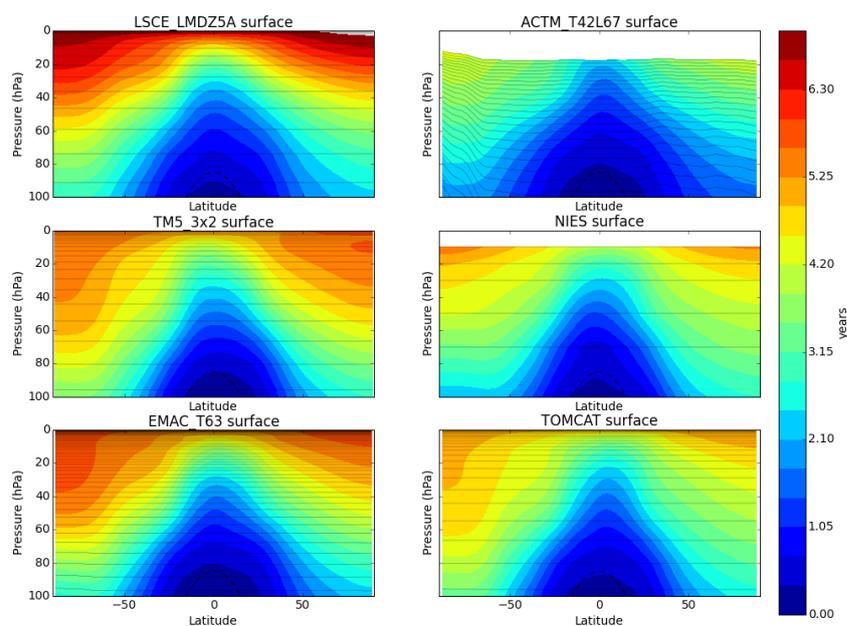


**Figure 6.** Latitude-longitude plot of the AoA (days) derived from tracer "Ocean" evaluated in the lowest model layer. The results have been averaged over the period 2000-2010 (11 years). Ages older than 60 days appear in dark red.

latitudes. Most notably, high altitude stations such as Niwot Ridge (3523 masl) and Mauna Loa (3397 m above sea level) show much smaller concentration differences with respect to the South Pole than e.g. Cape Kumukahi at sea level. This is confirmed by the middle panel, in which the modeled SF<sub>6</sub> fields are zonally averaged over the Pacific Ocean only (from 150°E to 210°E). In this average all models except NIES agree well within 0.1 pmol mol<sup>-1</sup> on the latitudinal gradient. Due to too reduced vertical mixing combined with fast latitudinal transport, the NIES model underestimates the latitudinal SF<sub>6</sub> gradient. The TOMCAT model simulates the observed latitudinal gradient of SF<sub>6</sub> on clean background stations well, but also shows high accumulation over land (see Figures 5 and 8, upper panel)

In Appendix A we further compare the simulated SF<sub>6</sub> latitudinal and vertical gradients to measurements made during the HIPPO campaigns (Wofsy, 2011).

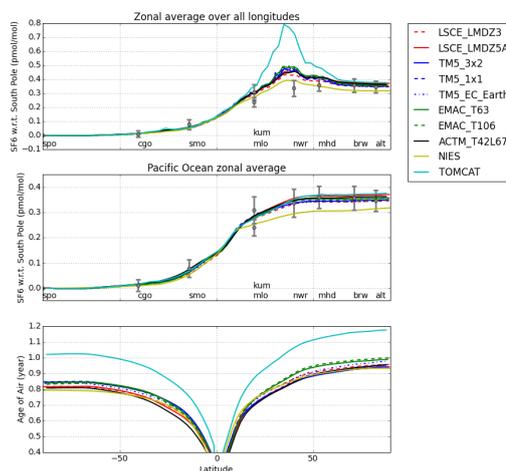
The lower panel of Figure 8 depicts a composite of the AoA of tracer "NHsurface" at the SH surface, and of tracer "SHsurface" at the NH surface. Again, all results are zonal averages over 2000–2010 (11 years) and include all longitudes in the zonal average. The lower panel confirms the NH–SH asymmetry noted earlier in Section 3.1, but also makes it clear that the asymmetry differs per model. Figure 9 highlights the NH–SH asymmetry by subtracting the "NHsurface" AoA sampled at the SH surface from the "SHsurface" AoA sampled at the NH surface. In all models, differences grow gradually from 10° latitude to the pole, where the AoA differences range between 0.10 year (TM5\_3x2) and 0.17 year (EMAC\_T106). The model ver-



**Figure 7.** Zonally averaged AoA (years) in the stratosphere for tracer "Surface". The results have been averaged over the period 2000-2010 (11 years). The thin black lines denote the mid-pressure levels of the model. For the ACTM model, this level is at around 17 hPa for the 50<sup>th</sup> layer. The dotted black line denotes the climatological tropopause. Note the linear y-scale that starts at 100 hPa.

sions with higher spatial resolution (TM5\_1x1, EMAC\_T106) show larger AoA differences compared to the lower resolution versions. The TM5\_EC-Earth version differs from the other TM5 versions, likely because the physics of the EC-Earth model (IFS version cy36r4) leads to differences in boundary layer mixing and convection, compared to the IFS model version that was used for the ERA-interim simulation (cy31r2).

- 5 From the Figure 8 (upper and lower panel) it is obvious that the TOMCAT model shows the strongest SF<sub>6</sub> gradient, along with the slowest IH transport as diagnosed from the tracers "NHsurface" and "SHsurface". NIES shows smaller SF<sub>6</sub> gradients, but AoA values in line with other models. Differences between the other models are less pronounced. The gradient of SF<sub>6</sub> is driven by emissions that take place mainly at midlatitude land masses in the NH. In contrast, tracers "NHsurface" and "SHsurface" are forced at the entire surface of both hemispheres. To make a meaningful comparison to the simulated latitudinal
- 10 gradients of SF<sub>6</sub>, we evaluate the AoA of tracers "NHsurface" and "SHsurface" at 50°S and 50°N respectively, and at the surface of the models. By adding these two ages, we align the resulting composite AoA with the bulk of the calculated latitudinal SF<sub>6</sub> gradient between the South Pole and 50°N in the middle panel of Figure 8. The result is shown in Figure 10. In this representation, the NIES model deviates by simulating weaker latitudinal SF<sub>6</sub> gradients. TOMCAT simulates a similar SF<sub>6</sub> gradient compared to other models, but differs in the composite AoA. Both models have weak vertical mixing (see Figure 2),

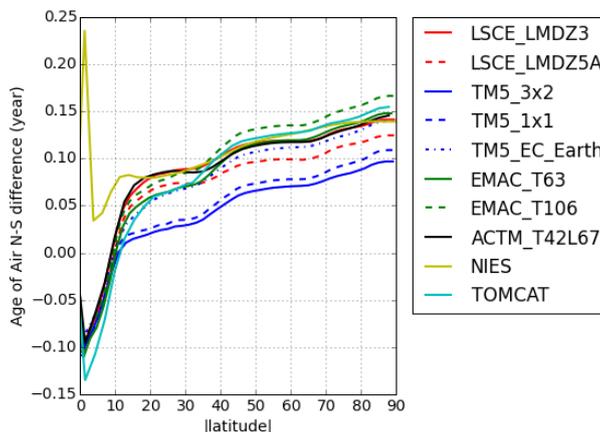


**Figure 8.** Upper two panels: Latitudinal gradient of SF<sub>6</sub> (pmol mol<sup>-1</sup>) at the surface, averaged over 2000–2010 (11 years). The upper panel includes all longitudes in the modeled zonal average. The middle panel includes only longitudes over the Pacific Ocean (150°E to 210°E). Modeled gradients have been scaled with respect to the South Pole. The grey symbols and their variability are calculated from the combined dataset constructed from flask data measured by the Halocarbons and other Atmospheric Trace Species (HATS) group from NOAA/ESRL and hourly CATS data (<https://www.esrl.noaa.gov/gmd/hats/combined/SF6.html>). Three letter codes at the bottom refer to the stations (alt = Alert, Canada; brw = Pt. Barrow, Alaska, USA; nwr = Niwot Ridge, Colorado, USA; brw = Pt. Barrow, Alaska, USA; kum = Cape Kumukahi, Hawaii, USA; mlo = Mauna Loa, Hawaii, USA; smo = Cape Matatula, American Samoa; cgo = Cape Grim, Tasmania, Australia; spo = South Pole; mhd = Mace Head, Ireland). Variability is calculated as the standard deviation of monthly time series of the station data relative to the South Pole station. Lower panel: Composite of the AoA (years) of tracer "NHsurface" at the SH and tracer "SHsurface" at the NH (see main text, averaged over 2000–2010).

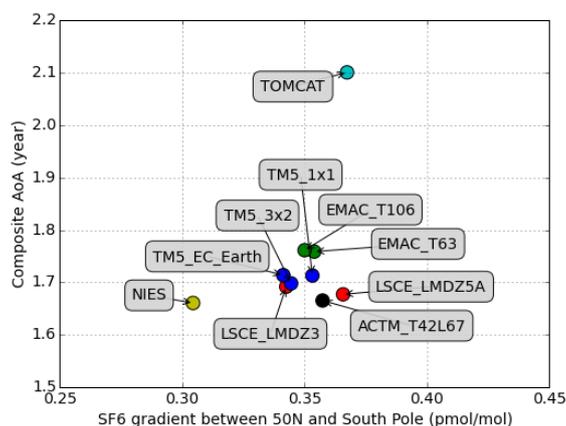
but NIES has fast horizontal transport, while TOMCAT accumulates SF<sub>6</sub> over the source regions over land (see upper panel in Figure 8). Different model versions cluster together in this representation, but differences between the LMDZ5A and LMDZ3 are larger. This illustrates the impact of the parameterisation of convective mixing in the LMDZ models (see Section 2.2).

### 3.5 Vertical transport

- 5 In this section, we focus on the vertical transport in the troposphere, as diagnosed from tracer "Land" and the simulated vertical gradients of <sup>222</sup>Rn. To this end, we show in Figure 11 (left-hand panel) the simulated mean vertical profiles, area weighted between 60°S and 60°N on a log-scale. Clearly, the TOMCAT and NIES models retain the <sup>222</sup>Rn closer to the surface, because of slower vertical mixing. The other models mix the <sup>222</sup>Rn to higher altitudes, but larger differences are found at pressures smaller than 400 hPa, where some models exhibit stronger increases at 150–300 hPa (e.g. ACTM, LMDZ5A), associated with
- 10 the parameterisation of convective mixing. The right-hand panel of Figure 11 shows the vertical profiles of the AoA of tracer "Land", averaged in the same manner.



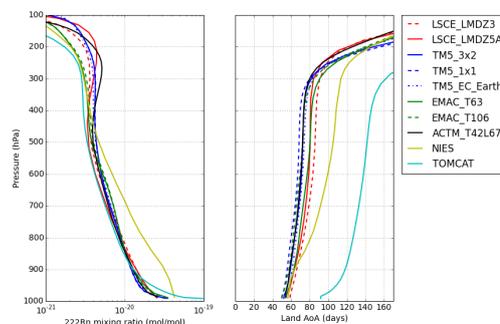
**Figure 9.** AoA difference (years) calculated from the values in the lower panel of Figure 8. The AoA at the SH of tracer "NHsurface" is subtracted from the AOA at the NH of tracer "SHsurface", and plotted against latitude. AoA is evaluated at the surface and averaged over 2000–2010 (11 years).



**Figure 10.** Composite AoA (years) plotted against the modelled SF<sub>6</sub> gradient. The composite AoA is calculated as the sum of the 50°N AoA of tracer "SHsurface" and the 50°S AoA of tracer "NHsurface". The SF<sub>6</sub> gradient is taken as the concentration difference between 50°N and the South Pole from the middle panel of Figure 8. Results are averaged over 2000–2010 (11 years).

To investigate consistent differences in vertical mixing between the models, Figure 12 compares the simulated vertical gradient in <sup>222</sup>Rn to the simulated AoA profiles of tracer "Land". Because <sup>222</sup>Rn decays radio-actively with a half-life of 3.8 days, we convert the vertical gradient of <sup>222</sup>Rn mixing ratio as:

$$\Delta = \ln^{222}Rn(950 \text{ hPa}) - \ln^{222}Rn(500 \text{ hPa}), \quad (3)$$

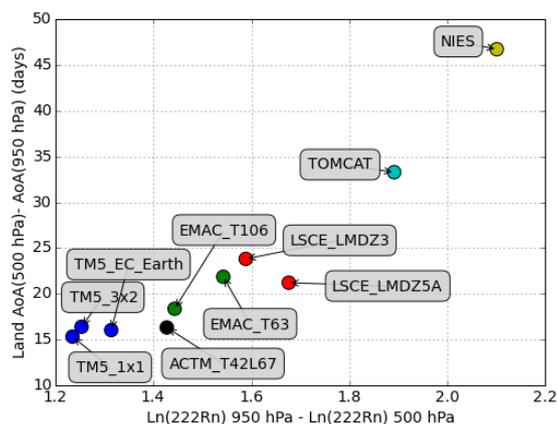


**Figure 11.** Vertical profiles of  $^{222}\text{Rn}$  (left,  $\text{mol mol}^{-1}$ , logarithmic axis) and the AoA of the "land" tracer (right, in days) simulated by the different models. The vertical profiles have been weighted-averaged between  $60^\circ\text{N}$  and  $60^\circ\text{S}$  and over the years 2000-2010 (11 years).

where we do not sample directly at the surface to avoid differences in sampling strategies between models (see Section 2). Here we quantify the gradient with respect to the 500 hPa level. In Figure 12 we plot the AoA gradient between 500 hPa and 950 hPa against  $\Delta$ . The results of the different models show a high degree of linear correlation, indicating that models with efficient vertical mixing (e.g. TM5, ACTM) have small AoA differences between the boundary layer and 500 hPa along with a flatter  $^{222}\text{Rn}$  profile. Models with slow vertical mixing (NIES, TOMCAT) show steeper  $^{222}\text{Rn}$  profiles along with larger  $\Delta$  values. Interestingly, the TM5 and EMAC models show that higher spatial resolution models lead to faster vertical mixing. This is different to the results found for IH transport and likely due to the fact that deep convective mixing is a sub-grid scale process. Numerics of such sub-grid scale processes are less prone to numerical diffusion compared to the advection process that drives the IH gradient. As expected, the two model versions of LMDZ that differ by their convective parameterisation show distinct differences in the vertical profiles.

### 3.6 Mapping the tropopause

Prather et al. (2011) introduced an idealised tracer (E90) to delineate the boundary between troposphere and stratosphere in transport models. Driven by surface emissions and a decay rate of  $90 \text{ days}^{-1}$ , tracer E90 is meant to quantify the rate of mixing of tropospheric air. Through numerical experiments, the tropopause in the UCI CTM (Prather et al., 2011) was defined as the surface on which the mixing ratio of E90 is  $90 \text{ nmol mol}^{-1}$ . Note that emissions of E90 are defined such that the global stationary state concentration of E90 is  $100 \text{ nmol mol}^{-1}$  (see Section 2). Mixing ratios in the stratosphere are lower, due to the long transport times. As alternative for E90, the AoA tracer "Surface" may also be used to delineate the stratosphere from the troposphere, as shown in Figure 2. In order to compare the tropopause pressure calculated by either E90 or the AoA surface tracer, we plot the tropopause pressure as a function of latitude in Figure 13. The tropopause pressure is calculated based on 2000–2010 zonal averages of E90 (unit  $\text{nmol mol}^{-1}$ ) and "Surface" AoA (unit days). For each latitude the tropopause pressure is determined by interpolation to  $\text{E90} = 90 \text{ nmol mol}^{-1}$  (dotted lines in Figure 13) and to "Surface" AoA = 90 days (solid lines in Figure 13). Results show a high level of consistency for these two metrics, although larger differences occur for several

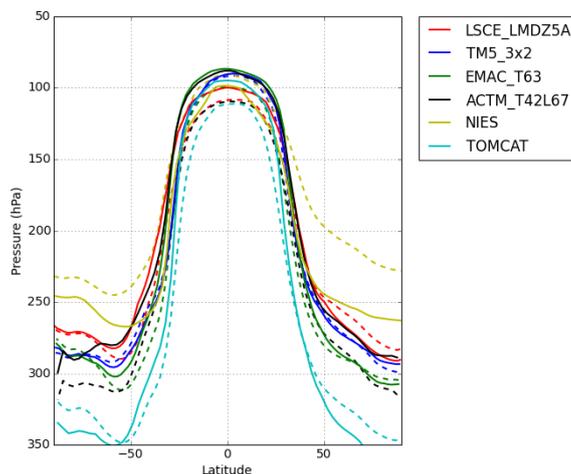


**Figure 12.** The 500 hPa - 950 hPa difference in AoA (days) of the "land" tracer plotted against  $\Delta$ : a measure for the vertical gradient in  $^{222}\text{Rn}$  (see main text). The vertical profiles used for the calculations are shown in Figure 11.

models. Differences in tropopause pressure based on E90 and tracer "Surface" are likely caused by the transport characteristics of the models. The "Surface" AoA simulation is forced by linearly increasing boundary conditions which are calculated back as AoA, while the E90 simulation is driven by surface emissions and a decay process with a 90-day turnover time. This causes different tracer gradients and hence difference in advective and convective transport. As a result, ACTM calculates systematically higher tropopause pressures for E90, while for NIES the reverse is observed. Other models show similar estimates for both tracers. When models are compared, both tracers calculate similar tropopause pressure differences, with NIES showing a very deep tropopause at higher latitudes and TOMCAT a shallower tropopause. Off-line models that use the same driver meteorological data (e.g. TOMCAT and TM5) would be expected to have a similar tropopause based on common temperature fields. However, since the tropopause pressures derived here depend on the transport of tracers by advection and convection, substantial differences are found that also depend on the choice of concentration (for tracer E90) and AoA (for tracer "Surface") at which the tropopause is evaluated. Finally, all models agree on a hemispheric asymmetric average tropopause with a tropopause pressure maximum around 55°S, likely associated with enhanced stratosphere–stratosphere exchange in the SH (Holton et al., 1995). In most models, this pressure maximum is more pronounced for tracer E90.

#### 4 Discussion

This TransCom AoA inter-comparison shows that the tropospheric AoA concept provides useful information on model transport characteristics. It was shown that the IH transport timescale in a particular model is strongly connected to the efficiency of vertical mixing and hence to the specific implementation of sub-grid-scale convective transport. Thus, the AoA metric may be used to better understand flux differences derived from CO<sub>2</sub> and CH<sub>4</sub> flux inversions (Patra et al., 2011; Law et al., 2008). The NIES model, which has slow convective mixing, still shows fast IH mixing, a small IH SF<sub>6</sub> gradient, and a deep extra-tropical



**Figure 13.** Zonal average tropopause pressure (averaged over 2000–2010) as a function of latitude, obtained from AoA tracer "surface" (solid lines) and E90 (dashed lines) for different models (see legend). Tropopause pressure is obtained from 1D linear interpolation to 90 days for AoA tracer "surface" and to 90 nmol mol<sup>-1</sup> for E90.

tropopause. In contrast, the TOMCAT model in the configuration used here combines weak vertical mixing with a stronger SF<sub>6</sub> gradient between land and ocean, and a shallower tropopause. The TM5 model, which was diagnosed with slow IH transport in the TransCom methane inter-comparison (Patra et al., 2011), now uses the convective fluxes stored in the ECMWF ERA-interim archive, which are based on Gregory et al. (2000) and other changes (Tsuruta et al., 2016; Dee et al., 2011). This brings the IH transport times close to other models (see Figure 10). This confirms the fact that the parameterisation of sub-grid convective fluxes in CTMs deserves attention, specifically when used in atmospheric inversion studies. This is in line with the study of Stephens et al. (2007), who showed that the attribution of the global CO<sub>2</sub> land sink depends strongly on the vertical mixing in models.

We found an interesting hemispheric asymmetry in the IH transport times, with "older" NH air diagnosed with an AoA tracer forced at the SH surface, compared to the age of SH air, diagnosed with an AoA tracer forced at the NH surface. We tentatively attribute this to the effect of land-masses in the NH, which lead to an atmospheric stabilisation in winter, decoupling the boundary layer (BL) from the overlying free troposphere (FT). This is partly compensated by more effective mixing in summer, but the resulting net effect is well known as the "seasonal rectifier" (Denning et al., 1995). Thus, the AoA metric can also be used to quantify the strength of these rectifier effects in CTMs. Another (related) factor that might be responsible for this asymmetry is the mean position of the Inter Tropical Convergence Zone (ITCZ) north of the equator (Schneider et al., 2014).

Another important timescale in GCMs and CTMs is the mixing time of the BL and its coupling with the overlying FT. Together with the aforementioned "convection" timescale, these two quantities seem responsible for the main model diversity in this inter-comparison. For instance, the TM5 model shows a relatively strong coupling between the BL and the FT, witnessed



by (i) relatively young air of the ocean AoA tracer over land (Figure 6), (ii) only a small IH difference in the AoA (Figure 9), and (iii) a small 950 hPa – 500 hPa vertical gradient in  $^{222}\text{Rn}$  and the land AoA tracer (Figure 12). Differences with the EMAC, LMDZ, and ACTM models seem to be determined by the strength of this BL–FT coupling. Differences with NIES and TOMCAT, however, seem to be driven by differences in the convective parameterisation.

5 Other model differences, such as resolution, advection scheme, the meteorological driver data, nudging, and the vertical coordinate system may also play a role and may become more apparent on the smaller spatial and temporal scales that are not part of this first analysis. In mapping the tropopause with either E90 or the AoA derived from the surface tracer (Figure 13) model-dependent differences appeared that may be related to the treatment of atmospheric transport. AoA tracers with linearly growing boundary conditions lead to different concentration gradients than tracers with surface emissions and first-order decay, such as E90. In regions with large gradients, such as the planetary boundary layer and the tropopause, differences in the numerical treatment of advection may lead to different transport characteristic of AoA tracers, compared to tracers with physical sources and sinks.

Our multi-model results agree qualitatively with the study of Waugh et al. (2013) that focused on  $\text{SF}_6$  and a AoA tracer in a single model (GMI-MERRA). They found "older" AoA than the age derived from  $\text{SF}_6$  observations. The AoA simulations presented here allow for more detailed analyses in the future, including comparisons to earlier efforts to quantify tropospheric AoA based on observations (e.g., Holzer and Waugh, 2015).

The simple AoA concept exploited here is easily implemented in CTMs, and other modellers are encouraged to implement AoA tracers in their CTM. The AoA protocol is also useful as benchmark for model development. Finally, the entire time series (1988–2015) may be used to study trends in the Brewer Dobson circulation (Fu et al., 2015) or IH transport timescales.

## 20 5 Conclusions

This paper presents the first results of the TransCom AoA inter-comparison with six participating models simulating five AoA tracers and four additional tracers over the time period 1988–2015. In this paper we mainly analysed averages over the 2000–2010 period (11 years). The main conclusions are:

- The IH transport time depends strongly on the strength of the convective parameterisation in the participating models.
- 25 – IH transport proceeds faster from the NH to the SH than from the SH to the NH. This is attributed to the seasonal rectifier effect caused by NH landmasses.
- BL mixing and venting to the FT over land varies among models, which leads to consistent differences in modeled vertical gradients.
- The AoA concept can be used to map the tropopause. At the tropopause the air has last been in contact with the surface 30 90 days ago (AoA = 90 days). For most models, the derived tropopause pressure is in good agreement with the E90 tracer (Prather et al., 2011).



- Upper stratospheric AoA ranges considerably among the participating models (4–7 years).

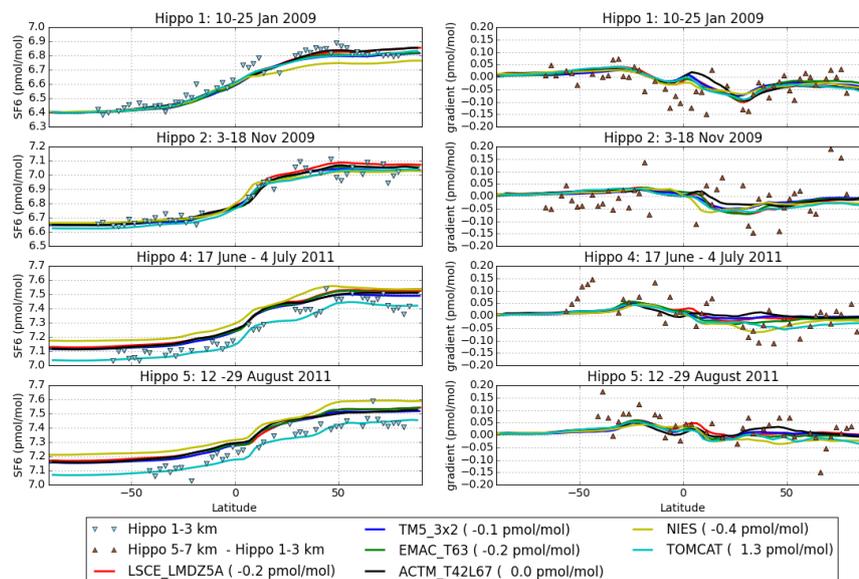
## 6 Code availability

Model output and analysis software in the form of a Jupyter Notebook (python) is available to the community. Apart from the AoA tracers discussed in this paper, also AoA tracers are available that are forced in the troposphere and stratosphere. These tracers are forced above and below a climatological pressure surface given by Lawrence et al. (2001). Results are not discussed, because not all models used the correct definition. Information on the availability of source code for the models featured in this paper is tabulated in Table A1.

### Appendix A: Comparison with HIPPO SF<sub>6</sub>

Figure A1 compares monthly mean SF<sub>6</sub> output from the models to observations made during the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observation (HIPPO) campaigns (Wofsy, 2011). Only HIPPO flights over the Pacific Ocean are included, and the corresponding model mixing ratios are averaged over the Pacific Ocean only (from 150°E to 210°E). Hippo 1 observations are compared to monthly means of January 2009, Hippo 2 to November 2009, Hippo 4 to June and July 2011 averages, and Hippo 5 to August 2011. The left panels show the latitudinal gradients averaged over 1–3 km. Since models used different SF<sub>6</sub> 1988 mixing ratios as initial fields and show different accumulation rates in the lower atmosphere, models have been shifted to match the 2009 observations. Note that the shifts (mentioned in the caption) range from +1.3 ppt for TOMCAT to –0.4 ppt for NIES. During the 2009–2011 Hippo period differences between these two models gradually increase further. This is likely related to fast accumulation in NIES due to limited vertical mixing, combined with fast horizontal transport in the lower atmosphere. TOMCAT was also diagnosed with slow vertical transport (see Figure 2) but also with slow horizontal mixing (signalled by "old" air over the ocean for AoA tracer "Land" in Figure 5). Other models agree on a faster increase of SF<sub>6</sub> in the models compared to observations, signalling a likely overestimate of the emissions in 2009–2011 (see Table 3). The latitudinal gradients are very similar among the models (except for NIES as in Figure 8) and generally agrees well with observations. The right-hand panels show the modeled and measured vertical SF<sub>6</sub> gradients, calculated by taken the difference between 5–7 km averages and 1–3 km averages, like in Patra et al. (2014). Modeled and measured vertical gradients are very small and measurement noise hampers drawing firm conclusions about the ability of models to correctly simulate SF<sub>6</sub> vertical gradients.

*Author contributions.* M.K. wrote the manuscript. M.d.B performed the TM5 simulations, L.K. the TM5\_EC-Earth simulations. H.O. and A.P. were responsible for EMAC, Y.Y., F.C., and P.B. for LMDZ, P.P for ACTM, D.B. and S.M. for NIES, and S.D, W.F, and M.P.C for TOMCAT. The protocol was initiated by M.K. and P.P.



**Figure A1.** Comparison of modeled monthly-mean SF<sub>6</sub> (averaged over the Pacific Ocean only: from 150°E to 210°E) to the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observation (HIPPO) campaigns (Wofsy, 2011), similar to comparisons presented in Patra et al. (2014). Left panels show models and observations averaged over 1–3 km altitude. Right panels are the gradient between 5–7 km averages and 1–3 km averages. Models have been shifted to match the HIPPO latitudinal gradients in 2009. Magnitudes of the shifts are indicated in the caption.

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

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**Table A1.** Availability of source code for the models featured in this paper.

Short Name	Code availability
LMDZ	LMDZ is open source free software under licence CeCILL ( <a href="http://www.cecill.info/licences/Licence_CeCILL_V2-en.html">http://www.cecill.info/licences/Licence_CeCILL_V2-en.html</a> ). The versions used here are available online via <a href="http://svn.lmd.jussieu.fr/LMDZ">http://svn.lmd.jussieu.fr/LMDZ</a> .
TM5	TM5 version control is performed on the SVN server of the Dutch Met Office (KNMI). The AoA simulations are performed using TM5-MP, revision r182, committed on 29-05-2017. Access to the SVN server is granted to researchers actively participating in model development.
TM5_EC-Earth	TM5_EC-Earth is the TM5-MP version incorporated in EC-Earth version 3.2.2. Information about the model and access to the code is available at <a href="http://www.ec-earth.org">http://www.ec-earth.org</a> . The specific version used for this study can be found under the branch r4353-Age_of-Air. This version consists of the main developmental version of EC-Earth updated until 04-07-2017, supplemented with the Age of Air code of TM5.
EMAC	The ECHAM/MESSy Atmospheric Chemistry (EMAC) model is a configuration of MESSy, which is being continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions that are members of the MESSy Consortium. Institutions can be a member of the MESSy Consortium by signing the Memorandum of Understanding. More information can be found on the MESSy Consortium website ( <a href="http://www.messy-interface.org">http://www.messy-interface.org</a> ).
ACTM	The ACTM code is not open source. The model code is based on CCSR/NIES/FRCGC AGCM5.7b. The copyright belongs to the developers at CCSR (Univ. Tokyo), NIES and FRCGC (JAMSTEC).
NIES	The NIES source code is proprietary. The model copyright is owned by developers at NIES.
TOMCAT	The TOMCAT code is not open source. The model is available to all UK researchers funded by the Natural Environment Research Council (NERC) and other UK research councils. The model is also available to other scientists with an interest in active collaboration with existing users, subject to limitations of resources to support the collaborations.

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