# Causal inference for ~~stratospheric chemistry: insights into~~ quantifying chemical–dynamical pathways controlling tropical middle stratospheric ozone variability

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**Abstract.**

~~This study investigates the coupling between chemicaland dynamical processes driving tropical middle stratospheric~~ Understanding the chemical–dynamical interactions controlling ozone ($O_3$) variability ~~using a~~ in the tropical middle stratosphere is essential for interpreting short-term trends and their sensitivity to dynamical fluctuations. This study applies a process-oriented causal inference framework that combines causal discovery ~~with~~ and causal effect estimation. This approach integrates qualitative physical knowledge through a causal graph applied to satellite observations and a chemistry-transport model (CTM) simulation~~. The analysis is split into two subperiods of monthly data : 2004–2011, characterized by an $O_3$ decrease, and 2012–2018, when $O_3$ increased in the tropical middle stratosphere~~, using monthly data for the period 2004-2021. Causal inference ~~identifies distinct processes governing $O_3$ behaviour. During 2004~~robustly identifies a dominant chemical–~~2011, a robust negative contemporaneous connection from~~ dynamical pathway, in which variability in residual vertical velocity ($w^*$) modulates nitrous oxide ($N_2O$~~to~~), subsequently affecting nitrogen dioxide ($NO_2$~~emerged, while in 2012–2018 this shifted to a one-month lag . This slower response reduced NO~~) and ultimately $O_3$. Direct causal effect estimates capture that $O_3$ variability is dominated by this indirect $NO_2$~~production from~~-mediated pathway, while the direct influence of $w^*$ on $O_3$ is weak. The total causal effect (direct and mediated) peaks at a lag of approximately two-three months, indicating that the response develops on this timescale as the influence propagates through $N_2O$ ~~oxidation, limiting $O_3$ loss via the $NO_x$ catalytic cycle. Further analysis across Quasi-Biennial Oscillation (QBO) regimes reveals regime-dependent differences in the causal links . The~~ and $NO_2$. Regime-oriented analysis applied to the observations reveals that the chemical links ($N_2O$~~to~~–$NO_2$ ~~connection is weaker during westerly shear, associated with reduced upwelling, and stronger during easterly shear, reflecting enhanced upwelling.~~ and $NO_2$–$O_3$) strengthen during westerly QBO shear compared to easterly shear.

Our study highlights the pivotal role that causal inference can play in disentangling complex chemical-dynamical influences on $O_3$, complementing traditional statistical methods. This approach lays the foundation for broader applications in stratospheric chemistry, where ~~many relations remain~~ the understanding of various feedback pathways remains uncertain. By discovering and quantifying causal links, this methodology ~~addresses~~ can be adapted to address open questions with environmental and societal relevance. Therefore, integrating causal reasoning into data-driven science enhances process understanding and also strengthens the synergy between machine learning and statistical methods in Earth and environmental sciences.

## 1 Introduction

Stratospheric ozone ($O_3$) is essential for protecting life on Earth by absorbing most of the harmful solar ultraviolet (UV-B) radiation (280–315 nm). The tropical ~~middle stratosphere~~ (10°S–10°N) middle stratosphere (∼10 hPa) is a key region for the photochemical formation of $O_3$ (Chapman, 1930). A balance between photochemical production and loss mainly determines the overall abundance of stratospheric $O_3$. Meanwhile, its global distribution and inter-annual changes are mainly determined by dynamical and chemical processes in conjunction with their superimposed variability of different origins and periodicity, such as Brewer–Dobson circulation (BDC), El Niño–Southern Oscillation (ENSO)~~and~~ , Quasi-Biennial Oscillation (QBO)~~. Depletion of~~ , concentrations of greenhouse gases (GHGs) and Ozone Depleting Substances (ODSs). Reduction in stratospheric $O_3$ concentrations causes more UV radiation to reach the Earth's surface, resulting in harmful effects, including damage to plant life and crops, disruption of aquatic ecosystems, and increased risks of skin cancer, cataracts, immune suppression, and erythema in humans (WMO, 2022; Zerefos et al., 2023). To mitigate these harmful effects, actions taken under the Montreal Protocol ~~(United Nations, 1987)~~ and its Amendments and Adjustments have significantly reduced emissions and atmospheric abundances of ~~ozone-depleting substances (ODSs)~~ODSs, contributing to the recovery of the stratospheric $O_3$ layer.

~~According to the recent Scientific Assessment of Ozone Depletion (WMO, 2022) and (Weber et al., 2022; Kuttippurath et al., 2024), aggregated ground- and space-based observations reveal that from 2000 to 2020, Total Column Ozone (TCO) in the tropical region did not show a significant positive trend (0.2 ± 0.3% per decade). This is due to contrasting vertical $O_3$ trends: stratospheric $O_3$ levels declined while tropospheric $O_3$ increased, both trends being unrelated to changes in ODS abundances (WMO, 2022). At the same time, recent studies have highlighted a complex structure in tropical~~

Apparent changes in tropical middle stratospheric $O_3$ ~~trends~~can vary substantially depending on the length and timing of the analyzed period. For example, ~~measurements from various satellite instruments consistently show increases in upper-stratospheric~~ in the early 2000s, several studies reported statistically significant $O_3$ ~~with a positive trend of ∼1–1.5% per decade, while a small though uncertain decline (1–2% ±5% per decade) has been observed in the lower stratosphere (WMO, 2022). The increase in upper-stratospheric~~ decline in this region using a variety of datasets and methodologies (Kyrölä et al., 2013; Eckert et al., 2014;
. Subsequent analyses showed that the slight differences in the tropical $O_3$ ~~results from reductions in ODSs alongside $CO_2$-driven cooling of the stratosphere. Conversely,~~ trends across different data sources arise from small shifts in the ~~lower-stratospheric decrease is linked to~~ analyzed time period, largely due to endpoint anomalies influenced by the ~~climate-change-driven acceleration of large-scale stratospheric circulation (Eyring et al., 2010; Dhomse et al., 2018). This is also supported by Thompson et al. (2021)~~

~~, who found that ozone trends in the tropical lower stratosphere become insignificant when accounting for tropopause height changes, indicating that chemical ozone loss is not a primary factor in these trends. However, recent work by Villamayor et al. (2023) suggested that halogenated very short-lived substances (VSLS) may account for about a quarter of the observed negative~~ curvature of long-term $O_3$ ~~trend in the tropical lower stratosphere during 1998–2018. Overall, the current understanding is~~ variability and unaccounted fluctuations in the record (Petropavlovskikh et al., 2019; Sofieva et al., 2021). Recent assessments show that the magnitude of $O_3$ trends in the tropical ~~stratosphere between around 25–40 km and over 2000–2020 are highly uncertain , with estimates ranging from -2 to 0% per decade across most datasets (WMO, 2022). Earlier studies focusing on the tropical middle (∼10 hPa) stratosphere but covering a slightly shorter period (2000-2016) did not reveal a definitive trend across various data sources (Petropavlovskikh et al., 2019; Steinbrecht et al., 2017; Godin-Beekmann et al., 2022). Recently, Arosio et al. (2024) also showed non-significant $O_3$ trends in the tropical middle stratosphere (2004-2021) below around 35 km based on satellite datasets and the simulations from TOMCAT, a three-dimensional (3-D) chemistry-transport model (CTM, see their Fig. 3). A complementary study by Szeląg et al. (2020) reports~~ middle stratosphere since the early 2000s is highly uncertain ( -2 to 0% per decade, WMO, 2022) with multiple studies finding no statistically significant or robust long-term trend across different datasets and time periods (Petropavlovskikh et al., 2019; Steinbrecht et al., 2017; Godin-Beekmann et al., 2022; Aros . In addition, Szeląg et al. (2020) highlighted a strong seasonal dependence in tropical middle stratospheric $O_3$ trends ~~from 2000 to 2018~~during 2000-2018, with a significant increase in spring (2-3% per decade) and a non-significant decrease in autumn (-1% to -2% per decade), resulting in ~~overall~~ non-significant trends~~when averaged across all seasons, which is in agreement~~ , consistent with Galytska et al. (2019); Li et al. (2023). ~~Considering these findings, it is important to note that the slight differences in the tropical $O_3$ trends across different data sources, arising from small shifts in the analyzed time period, can be attributed to endpoint anomalies influenced by the curvature of~~ However, the absence of a persistent long-term trend in recent analyses does not rule out future changes of $O_3$ ~~variability and unaccounted fluctuations in the record (Petropavlovskikh et al., 2019; Sofieva et al., 2021). The studies mentioned above do not reveal pronounced $O_3$ trends~~ in the tropical middle stratosphere~~over extended periods (exceeding approximately 15–20 years), with minor shifts varying by a few years in different directions. This is in contrast to earlier studies reporting a significant decline in $O_3$ in the early~~ . Similarly, negative trends observed in the early 2000s ~~. For example, Kyrölä et al. (2013) and Gebhardt et al. (2014) were among the first to detect negative $O_3$ changes through the analysis of different datasets. Further studies (Eckert et al., 2014; Nedoluha et al., 2015; Galytska used a variety of datasets and methodologies to confirm the presence of negative $O_3$ trends . Ball et al. (2018) attributed the observed decrease in tropical middle stratospheric $O_3$ primarily to high $O_3$ levels during the 2000~~could recur if comparable dynamical–~~2003 period. However, one might question the relevance of the~~ chemical conditions arise. In this context, understanding the mechanisms governing interannual $O_3$ ~~decline observed during the early 2000s, given its occurrence over a relatively short time frame and the absence of a similar trend in analyses of the longer periods within the past 20 years. Nonetheless, understanding~~ variability becomes particularly important, as such variability can substantially influence trends derived from a limited observational dataset.

While previously discussed $O_3$ trends motivate this study, our objective is to quantify the mechanisms that ~~influence~~ control $O_3$ ~~behavior remains essential, as past trends do not preclude the possibility of their recurrence.~~

~~To explore the causes of the observed~~ variability on monthly timescales and thus, that can modulate trends over limited time periods. The sensitivity of $O_3$ ~~decline~~ trends in the tropical middle stratosphere ~~, Galytska et al. (2019) demonstrated that it was ultimately~~ to the analyzed period highlights the dominant role of chemical–dynamical variability in this region. In this context, Galytska et al. (2019) showed that the $O_3$ decline observed in the early 2000s was dynamically controlled and ~~occurred due~~ linked to increases in nitrogen dioxide ($NO_2$). The elevated $NO_2$ levels enhanced $O_3$ loss through the catalytic $NO_x$ ($NO_x$ = NO + $NO_2$) cycle, dominant in the middle stratosphere (Portmann et al., 2012). The increase in $NO_2$, also confirmed by Dubé et al. (2020), resulted from the prolonged residence time of its primary source, nitrous oxide ($N_2O$). ~~Anthropogenic $N_2O$ is not regulated under the Montreal Protocol (WMO, 2022) and is only addressed by the Kyoto Protocol to the United Nations Framework Convention on Climate Change (UNFCCC). Controlling $N_2O$ emissions would make a major contribution to mitigating long-term climate effects (Nisbet et al., 2021) and stratospheric $O_3$ depletion (Ravishankara et al., 2009). Over the past 40 years, the rise in atmospheric $N_2O$ has been primarily driven by human-induced emissions, with agricultural sources such as the use of N-fertilizers being the main contributor (Weber et al., 2024). The ongoing increasing levels of $N_2O$ will lead to enhanced $NO_x$-catalysed $O_3$ depletion in the middle atmosphere (Chipperfield and Bekki, 2024).~~ Since $N_2O$ is a long-lived species (Portmann et al., 2012; Chipperfield et al., 2014), the changes in its abundance reflect variations in tropical upwelling within the BDC, therefore, they are directly affected by changes in stratospheric transport (Iglesias-Suarez et al., 2021). Prather et al. (2023) showed that during 2005-2021, $N_2O$ increased in the tropical middle stratosphere more than expected from the rate of tropospheric increases. This ~~implied~~ implies a more vigorous BDC, which leads to a shorter lifetime. Minganti et al. (2020) evaluated the climatological impact of the stratospheric BDC on $N_2O$ and reported that, in the tropical middle stratosphere, the interannual variability of vertical residual advection exhibits a significant spread, which reflects the influence of the major source of interannual variability in the equatorial stratosphere, i.e. the QBO on tropical upwelling (Abalos et al., 2015). Chipperfield and Gray (1992), and later Park et al. (2017) highlighted that the source gas $N_2O$ and the reactive nitrogen species ($NO_y$) display coherent QBO signals within the tropical stratosphere. $O_3$ also exhibits a strong QBO signal, with the QBO directly influencing $O_3$ levels by altering the chemical reactions responsible for $O_3$ depletion, see also Chipperfield et al. (1994) and Ming et al. (2025). This oscillation modulates the rates of these reactions, leading to additional variations in $O_3$ concentration.

~~**Overview of $O_3$ variability in the tropical (10°S-10°N) middle (10 hPa) stratosphere.** (a) Annual mean anomalies of $O_3$ for the period 2002-2018. Solid lines represent the sources of the long-term records: OSIRIS (blue), SWOOSH (grey), SAGE-II-SCIAMACHY-OMPS (green), and TOMCAT CTM (orange), with the shaded regions indicating the 95% confidence intervals of the changes. Dashed lines represent simple linear fits for the subperiods 2002-2011 and 2012-2018. (b) Scatter plots of $w^*$ monthly anomalies from reanalysis and $N_2O$ monthly anomalies from observations (circles) and the TOMCAT simulation (triangles), split into the periods 2004-2011 (orange) and 2012-2018 (pink). (c) Same as (b) but for $N_2O$ and $NO_2$. (d) Same as (b) but for $NO_2$ and $O_3$. The Pearson correlations (r) for observations and TOMCAT are indicated above the panels (b–d) for both time periods.~~

~~To~~ Therefore, to set the stage for the ~~analysis of the complex~~ subsequent causal analysis of these chemical-dynamical ~~influences~~ feedback pathways on $O_3$ variability ~~during 2004-2018, we focus on the deep tropical (10°S-10°N) middle stratosphere(10~~

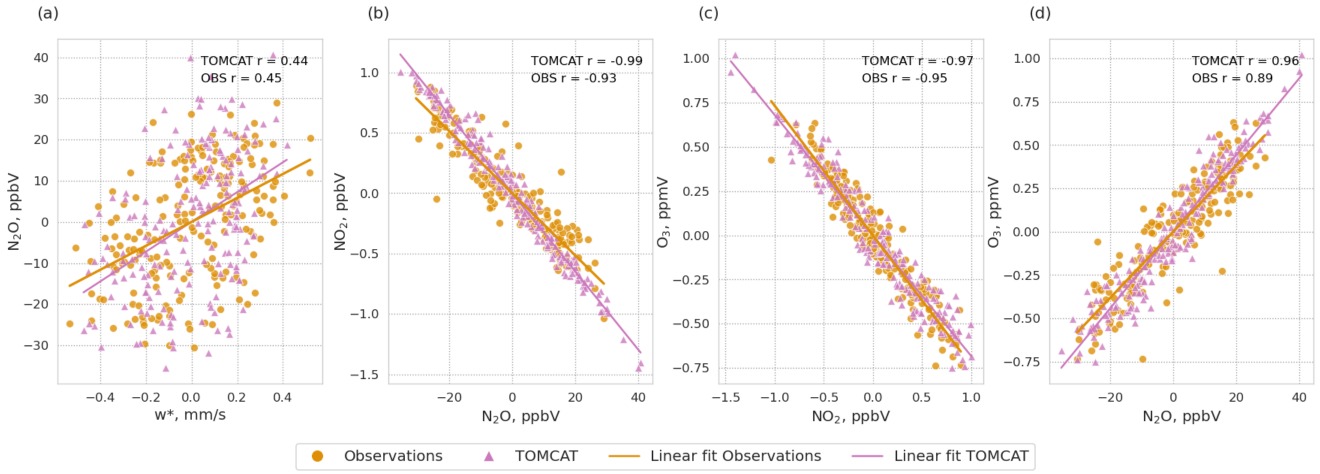

**Figure 1.** Scatter plots of detrended monthly mean anomalies in the tropical middle stratosphere for 2004–2021, from observations (circles) and the TOMCAT CTM simulation (triangles). Panels show (a) $w^*$ versus $N_2O$, (b) $N_2O$ versus $NO_2$, (c) $NO_2$ versus $O_3$, and (d) $N_2O$ versus $O_3$. Solid lines indicate linear regressions for observations (orange) and TOMCAT (pink). The corresponding Pearson correlation coefficients ($r$) are shown in each panel.

~~hPa) region. Figure 1 a illustrates the annual mean~~ in the tropical middle stratosphere, Figure 1 presents scatter plots of detrended monthly mean anomalies of (a) residual vertical velocity ($w^*$) versus $N_2O$, (b) $N_2O$ versus $NO_2$, (c) $NO_2$ versus O~~$_3$ anomalies divided into two distinct periods: 2002–2011, characterized by adecline in $O_3$levels (Kyrölä et al., 2013; Gebhardt et al., 2014; E~~ ~~, and 2012–2018, marked by increasing O~~, and (d) $N_2O$ versus $O_3$ ~~levels (Arosio et al., 2019). The analyzed data agree on an overall $O_3$ decline during 2002-2011 and an increase in the following 2012-2018 period. These contrasting $O_3$ changes over the two subperiods serve as motivation for the subsequent causal analysis (see Sect. 3) of the processes driving chemical-dynamical interactions in the tropical middle stratosphere. The bi-annual variability in the observed O $_3$ anomaly time series (Fig. 1a) is associated with the QBO signal. Figure 1b-d shows scatter plots of monthly anomalies~~ for observations and ~~a~~ the TOMCAT CTM simulation for ~~the analyzed subperiods defined in Fig. 1a. Note that the first subperiod is 2004-2011, as $N_2O$ data is not available before this period.~~ 2004-2021. Both observations and the TOMCAT ~~simulations~~ CTM simulation exhibit similar correlations and slopes. The relationship between ~~residual vertical velocity ($w^*$ )~~ and $N_2O$ is moderately positive (panel ~~b~~a). The strong ~~negative correlations~~ anti-correlations between $N_2O$ and $NO_2$ (panel ~~c) indicate that~~ b) result from their opposing response to transport-driven variability, where, e.g. enhanced upwelling increases $N_2O$ ~~is the major source of~~ while reducing its chemical loss, which is crucial for $NO_2$ ~~, and~~ production (see further discussion in Sect. 4.1). The negative relationship between $NO_2$ and $O_3$ (panel ~~d~~c) is a result of $NO_2$ being the primary sink of $O_3$ in the middle stratosphere (Park et al., 2017). The positive relationship between $N_2O$ and $O_3$ (panel d) is consistent with both tracers exhibiting lifetimes that exceed their vertical transport timescales in this region (Bönisch et al., 2011).

While the chemical–dynamical coupling governing tropical middle-stratospheric $O_3$ is understood, previous research relies mostly on correlation and different types of regression analyses that do not explicitly distinguish between direct and mediated effects within a multivariate system. A causal inference framework proposed in this study represents the relationships between the selected variables as a directed acyclic graph (DAG), in which nodes correspond to physical quantities and edges represent causal influences. Therefore, the structure of the DAG is informed by established physical understanding and subsequently evaluated using causal inference methods applied to the data. Causal inference can then be used to estimate the targeted relationships under explicit assumptions. Applying such an approach to a well-understood chemical–dynamical system provides a reliable test of whether data-driven causal inference can recover known physical pathways and quantify their contributions in the analyzed system. It is important to highlight that to ensure statistical consistency, this study analyzes detrended anomalies rather than long-term trends. Therefore, the results describe variability-driven processes and should not be interpreted as a direct explanation of decadal $O_3$ trends caused by externally forced long-term changes in $CO_2$, $N_2O$ emissions, or ODSs, but rather in the context of dynamical processes that are strongly influenced by the QBO.

## 2 Data sources

This study uses monthly data from satellite observations, reanalysis products, and a TOMCAT CTM simulation, ~~with a focus on the following key variables:~~ focusing on four core variables, namely $w^*$, $N_2O$, $NO_2$, and $O_3$. We also use the QBO as additional variables to examine how the relationships among the four core variables vary under different thermodynamical regimes. By selecting these variables, we focus the analysis on the interpretability of key chemical-dynamical processes that play a major role in controlling $O_3$ in the tropical middle stratosphere. We intentionally limit the number of variables to maintain a high level of interpretability of the causal graphs. We ~~analyze~~ then later compare the causal graphs from ~~different sources, namely~~ observations and the TOMCAT CTM simulation.

### 2.1 Observations

Since no single satellite instrument provides all the required variables with sufficient temporal and spatial resolution, we integrate data from multiple sources into this study. The following observational or reanalysis-based datasets were used:

- $w^*$: Derived Transformed Eulerian Mean (TEM) momentum terms (v0.1.1, Serva, 2022), based on ~~ECMWF~~ European Centre for Medium-Range Weather Forecasts (ECMWF) ERA5 reanalysis (Hersbach et al., 2020) using diagnostics from ~~Serva (2023)~~Serva (2023); Serva et al. (2024);
- **$N_2O$**: Profiles from the Earth Observing System (EOS) Microwave Limb Sounder (MLS~~)~~, v5.01) instrument on NASA's Aura satellite, which offer a vertical resolution of 5–8 km and a horizontal along-track resolution of 165–265 km (Lambert et al., 2020);

- **$NO_2$**: Profiles retrieved from limb-scattered sunlight observations ~~from~~ on the OSIRIS instrument aboard the Swedish Odin satellite (Murtagh et al., 2002; Llewellyn et al., 2004). OSIRIS $NO_2$ v7.3 is retrieved via spectral fitting in the 435–477 nm range from 10.5 to 39.5 km with a 2-3 km vertical resolution in most of the stratosphere (Dubé et al., 2022). Due to the pronounced diurnal cycle of $NO_2$ (Galytska, 2019; Dubé et al., 2020) the photochemical correction from Dubé et al. (2020) was applied to standardize all measurements to a reference time of 12:00 p.m.;

- **$O_3$**: OSIRIS $O_3$ v7.3 (Bognar et al., 2022), an improved version of the v5.10 product with corrected long-term drift by accounting for systematic errors in the instrument limb-pointing (Bourassa et al., 2018)~~. SWOOSH v2.7 data (Davis et al., 2016) and merged SAGEII-SCIAMACHY-Ozone Mapping and Profiler Suite (OMPS) (Arosio et al., 2018) were used to illustrate ozone trends in the tropical middle stratosphere over various subperiods in the Introduction in Fig. 1a. Additionally, we used equatorial QBO wind data~~ ;

- **QBO**: equatorial 10 zonal mean zonal wind taken from the Institute of Meteorology and Climate Research (KIT)~~for process-oriented analysis based on different QBO phases in Fig. ??.~~ , see

~~The combination of ERA5 $w^*$, MLS $N_2O$, and OSIRIS $NO_2$ and $O_3$~~ In the following, the combination of these datasets is collectively referred to as "observations".

### 2.2 TOMCAT Chemical Transport Model

TOMCAT is a three-dimensional off-line CTM (Chipperfield, 2006), driven here by winds and temperatures from the ERA5 reanalysis (Hersbach et al., 2020). Given prescribed atmospheric transport and temperature fields, TOMCAT calculates the distributions of chemical species in the troposphere and stratosphere. A stratospheric full-chemistry simulation, including all of the $NO_y$ chemistry discussed in this paper, was run at a horizontal resolution of 2.8° × 2.8° with approximately 1.5 km vertical resolution in the stratosphere (Chrysanthou et al., 2025). The model uses time varying sulfate aerosol surface area density Dhomse et al. (2015), solar fluxes (Dhomse et al., 2016), and lower boundary concentrations of GHGs and ODSs (WMO, 2022), recommended for CMIP6 simulations. We used the monthly average output in our analysis. The TOMCAT CTM simulation was chosen for its ability to provide a continuous time series without spatial or temporal gaps, making it ideal for robust comparison with the observational datasets.

### 2.3 Data preprocessing

Monthly anomalies ~~were calculated with respect to the 2004–2018 climatology~~ of $w^*$, $N_2O$, $NO_2$, and $O_3$ in the tropical (10°S–10°N) middle (10 hPa) stratosphere ~~for selected variables, namely $w^*$, $N_2O$, $NO_2$ and $O_3$~~ were calculated from observations and the TOMCAT CTM simulation ~~.~~ relative to the climatological mean over August 2004-December 2021. The analysis begins in August 2004, due to the availability of MLS $N_2O$ observations. However, for simplicity, we refer to the period as 2004-2021. To maintain a robust comparison against temporal sampling discrepancies, TOMCAT CTM data were masked to align with observations, omitting any dates with missing observational data. For causal inference, all time series were standardized. Since the application of causality requires the stationarity of the time series (see Sect. 3.1), we remove the linear trend from all analyzed time series. For more details about preprocessed timeseries from observations and the TOMCAT CTM simulation and their further comparison, see Appendix A.

## 3 Methods

### 3.1 Causal inference

We apply the latent Peter-Clark momentary conditional independence (LPCMCI) algorithm (Gerhardus and Runge, 2020), which is an extension of the PCMCI+ algorithm specifically designed to deal with latent (i.e. unobserved) variables (Runge et al., 2019; Runge, 2020). LPCMCI employs ideas from the Fast Causal Inference (FCI) algorithm to learn not only directed causal relationships but also infer the presence of latent confounders (Spirtes, 1995). LPCMCI benefits from the same ideas underlying PCMCI+ by increasing the effect size of conditional independence (CI) tests through including causal parents in conditioning sets. The LPCMCI method seeks to learn a Directed Partially Ancestral Graph (DPAG), which captures the causal relationships among the observed variables. Contrary to Maximal Ancestral Graphs (MAGs) that contain directed arrows ($\rightarrow$) and bidirected edges (in other words, double-headed arrows $\leftrightarrow$), PAGs can include additional edge types. These edges, drawn as $\circ\!\!\rightarrow$ and/or $\circ$-$\circ$, indicate the presence of hidden variables or uncertainty about the exact causal direction.

To understand the causal structure of the underlying complex dynamical system, the observed time series $\mathbf{X_t} = (X_t^1, ..., X_t^N)$, where $N$ stands for the different variables represented by time series, is assumed to follow the following causal process:

$$X_t^j = f_j\left(\mathcal{P}(X_t^j), \eta_t^j\right),$$

where $f_j$ is a measurable function that depends on all its inputs, $\eta_t^j$ represents dynamical noise (independent across $t' \neq t$). Here, $\mathcal{P}(X_t^j) \subset \mathbf{X}_{t+1}^- = (\mathbf{X}_t, \mathbf{X}_{t-1}, \ldots) \setminus \{X_t^j\}$ denotes the set of parent variables of $X_t^j$ since the value of $X_t^j$ is determined from the variables in $\mathcal{P}(X_t^j)$ and the dynamical noise $\eta_t^j$. Bidirected links between $X_t^i$ and $X_t^j$ in this model imply that the associated noise terms $\eta_t^i$ and $\eta_t^j$ are dependent through unobserved confounding. We assume causal stationarity, meaning $X_{t-\tau}^i \in \mathcal{P}(X_t^j)$ if and only if $X_{t-\tau-\Delta t}^i \in \mathcal{P}(X_{t-\Delta t}^j)$. In practice, causal discovery algorithms~~such as LPCMCI~~, including LPCMCI, require approximately stationary time series~~to~~, meaning time series whose statistical properties, such as mean and variance, remain approximately constant over time. Nonstationary behavior, such as long-term trends, can introduce spurious statistical dependencies and bias CI tests, leading to incorrect causal links. Therefore, removing or accounting for such trends (via masking or sliding window) is a methodological necessity to ensure that the ~~estimated conditional dependencies are not biased by nonstationary trends.We~~ algorithm identifies causal relationships associated with the internal dynamics of the system rather than coincidental alignment of long-term shifts in the variables (Runge et al., 2019, 2023).

Additionally, we also assume the absence of cyclic causal relationships, which, due to the temporal order, limits interactions to contemporaneous cases only when $\tau = 0$. Furthermore, we assume the standard assumptions of constraint-based causal discovery, the Markov condition and the faithfulness condition (Spirtes et al., 2000; Runge et al., 2023), which implies that conditional independence in the observed distribution generated by the ~~SCM above implies d-separation~~ structural causal model above implies directional separation (i.e. separation of variables by conditioning on appropriate sets of other variables) in the associated time series graph and vice versa.

We conducted a series of sensitivity tests with various settings of $\alpha_{pc}$ and $\tau_{max}$, but only used the causal graphs ~~for~~ with $\alpha_{pc}$=0.02 and $\tau_{max}$ = 1 for the toy model (see Sect. 4.1), ~~observations,~~ and with $\alpha_{pc}$=0.05 and $\tau_{max}$ = 2 for the observations and the TOMCAT CTM simulation (see Sect. 4.2). Since some of the analyzed variables have non-gaussian distributions, we use the RobustParCorr conditional independence test, which transforms the data to a normal distribution before the partial correlation test. This usage implies that we assume the functions $f_j$ in the model above to be linear. As a trade-off to its ability to also deal with latent confounding, LPCMCI suffers from lower recall compared to, e.g., PCMCI+ (Runge, 2020). Despite this, LPCMCI successfully identified causal connections in observational data that align closely with expert knowledge and the literature review. ~~Note, however, that we included one link assumption in this study, namely that the residual vertical velocity $w^*$ does not directly influence $O_3$. For the process-oriented analysis (Sect. ??) we further assumed that $N_2O$ has no direct influence on $O_3$. These links were left out of the initialization of the LPCMCI method.~~ However, LPCMCI did not robustly detect anticipated connections in the TOMCAT CTM simulation. To estimate the direct causal effects for the ~~three (sub)periods~~ period 2004-2021 in the TOMCAT CTM simulation, we refined the causal graphs by incorporating expert knowledge and insights from the literature review (as further discussed in Sect. 3.3 and depicted in Fig. 2 ).

### 3.2 Causal effect estimation

Over a hundred years ago, Wright (1921) suggested a method to estimate causal effects in linear models. This approach estimates the so-called path coefficients for all links in causal paths and then sums the products of these path coefficients over all causal paths. This method applies only to ~~directed acyclic graphs (DAGs)~~DAGs; therefore, in the case of DPAG, we ensure that all edges are directed before applying causal effect estimation. Causal effect estimation consists of the following steps:

1. For all causal links $i \to j$ that belong to causal paths from $X$ to $Y$ (where $X$ is a parent and $Y$ is a child), estimate the path coefficient $\beta_{i\to j}$ by regressing $j$ on all its parents and taking the coefficient corresponding to parent $i$. 2. The causal effect is then computed as:

$$CE = \sum_{\text{causal paths}} \prod_{\text{link } i\to j \text{ in path}} \beta_{i\to j}$$

By restricting this estimator to paths that pass through at least one node among a selected set of mediators $M^*$, it is also possible to compute mediated causal effects (MCE). These are defined as:

$$MCE = \sum_{\text{causal paths through at least one } M\in M^*} \prod_{\text{link } i\to j \text{ in path}} \beta_{i\to j}$$

### 3.3 Process-oriented causal inference framework

~~To advance the understanding of~~

To quantitatively characterize $O_3$ variability in the tropical middle stratosphere during ~~the 2004-2018 period and the two subperiods defined in Fig. 1a (2004-2011 and 2012-2018)~~2004–2021, we employ a process-oriented causal inference approach depicted in Fig. 2. Although causal analysis has already gained significant application in atmospheric sciences, including the analysis of Arctic processes and their links to middle latitudes (Polkova et al., 2021; Docquier et al., 2022; Galytska et al., 2023; Kretschmer et al., 2020), teleconnections (Karmouche et al., 2023; Tibau et al., 2022; Carvalho-Oliveira et al., 2024), atmosphere-biosphere interactions (Krich et al., 2020), and evaluating climate models (Nowack et al., 2020; Debeire et al., 2025) and their sensitivities (Ricard et al., 2024), it has not yet been applied to the study of stratospheric chemical-dynamical interactions.

Figure 2 outlines the process-oriented causal inference framework, which is built upon three essential components (blue squares): (1) a causal graph that contains information about qualitative cause-and-effect relationships (Runge et al., 2023), (2) observational and/or modeled data, and (3) a method for estimating causal effects. To construct (1) the causal graph, we employ a triangulated approach (Denzin, 2010; Uleman et al., 2024) that integrates (i) expert knowledge, (ii) a comprehensive literature review, and (iii) a data-driven causal discovery algorithm. While each of these components can independently contribute to the creation of the causal graph, we recommend employing the triangulated approach to ensure a more robust and reliable framework. To assess the performance of the selected (iii) causal discovery algorithm before applying it to real-world data, we first construct a (iv) "toy model" using synthetic data. This synthetic dataset is designed to replicate the properties and challenges of the real system while incorporating known underlying ground truth from (i) expert knowledge and (ii) a literature review. The toy model is then used to evaluate the performance of the causal discovery method in realistic, finite sample scenarios (Camps-Valls et al., 2023). To ensure the robustness of the results, it is recommended to further perform sensitivity tests on free algorithm parameters, such as $\alpha_{pc}$ and $\tau_{max}$ for (iii) the causal discovery algorithm and (iv) the toy model.

It is important to note that if (iii) the causal discovery algorithm does not robustly detect anticipated relationships in the analyzed (2) real-world or modelled data, the causal graph may be constrained based on (i) expert knowledge and (ii) a comprehensive literature review, including previous successful applications of causal discovery to related research topics. The final (1) causal graph, derived from (2) real-world data, serves as a foundation for estimating (3) direct and total causal effects. Causal effect estimation can be further refined through a process-oriented analysis, such as, for example, masking on different atmospheric regimes. Additionally, (v) total causal effects can be assessed across different time lags. This complex approach outlined in Fig. 2 ensures robust and reliable causal inference, particularly for complex systems such as the stratospheric chemical-dynamical interactions investigated here.

### 3.4 Confidence intervals and masking for regime-oriented analysis

Confidence intervals for direct causal effect estimates, computed using Wright's path coefficient (Sect. 4.2), were obtained via bootstrapping with 500 members. Only significant direct causal effects are shown, defined as when the bootstrap confidence interval does not include 0. For both direct and total causal effects across different time lags (Sect. 4.4), confidence intervals were likewise obtained from 500-member bootstrapping.

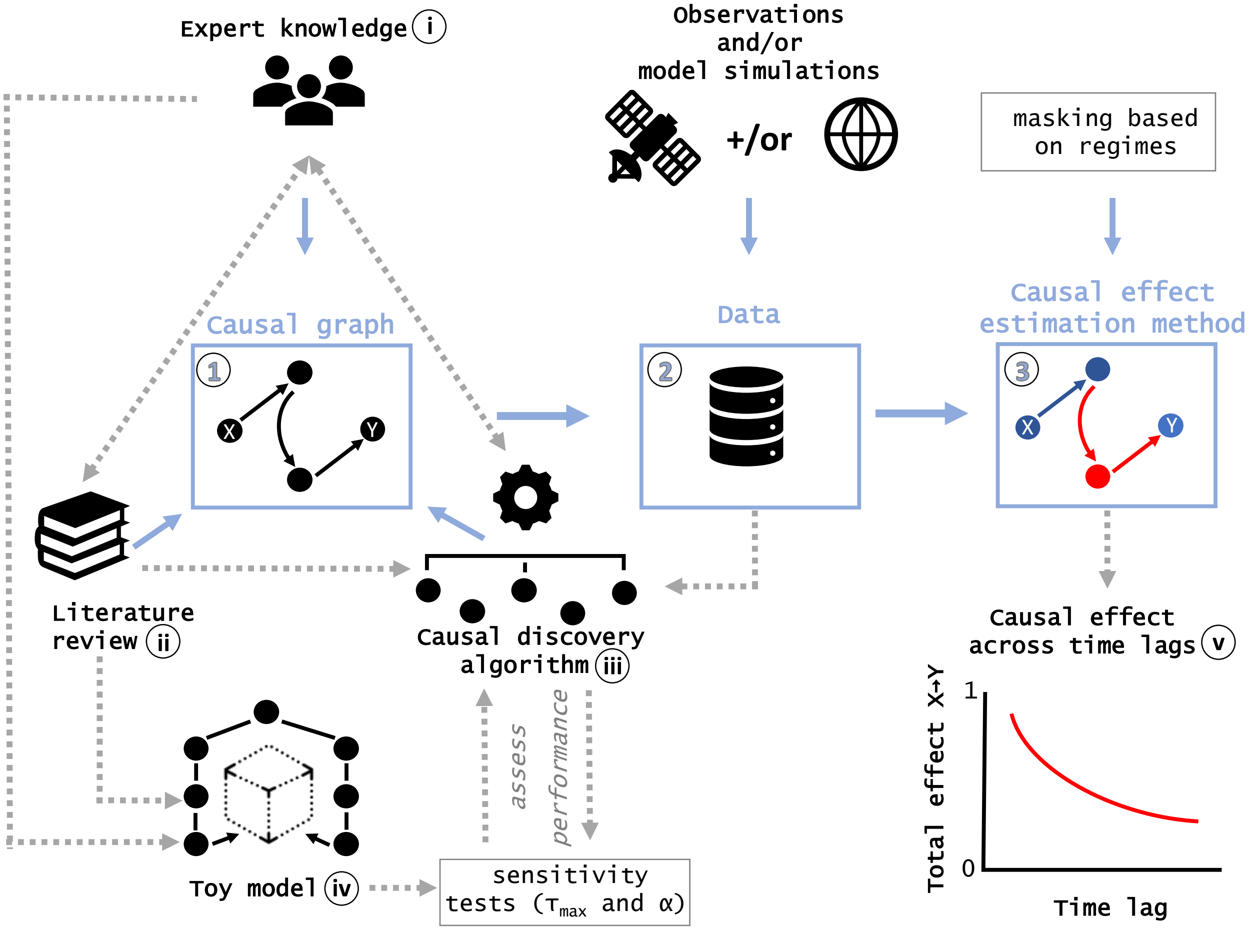

**Figure 2.** Process-oriented causal inference framework built upon three essential components (blue squares): (1) a causal graph, (2) data, and (3) a method for causal effect estimation. To construct the (1) causal graph for the studied system, a triangulated approach (Uleman et al., 2024) is applied, integrating (i) expert knowledge, (ii) a literature review, and (iii) a data-driven causal discovery algorithm. Before applying the (iii) causal discovery algorithm to real-world data, we construct a (iv) toy model to assess the performance of the selected (iii) causal discovery algorithm. The final (1) causal graph, based on (2) real-world data, serves as a foundation for estimating (3) causal effects, which can be further refined through process-oriented analysis, including masking on atmospheric regimes and additional sensitivity tests.

For the regime-oriented analysis of causal effects during different QBO phases (Sect. ~~??~~4.3), we first calculate the QBO wind shear as the ~~zonal wind gradient~~ vertical gradient of the zonal mean zonal wind between 10 ~~hPa~~ and ~~12 hPain log-pressure coordinates~~30 hPa. For observations, the shear is derived directly from radiosonde-based zonal wind at the two pressure levels (Kerzenmacher and Braesicke, 2026). For the TOMCAT CTM simulation, the zonal wind is first averaged over 10°S–10°N before computing the vertical gradient between 10 and 30 hPa. The resulting shear time series is subsequently standardized for use in the process-oriented causal analysis. Positive (negative) values correspond to a westerly (easterly) shear zone, which plays a ~~central~~ key role in modulating secondary circulation and stratospheric transport. We focus on the ~~10-12~~ 10-30 hPa shear layer since no data is available above 10 hPa in the used observational record.

## 4 Results and Discussions

### 4.1 Causal justification and validation with a toy model

Before applying causal discovery to ~~investigate~~ analyze chemical–dynamical interactions ~~in the tropical middle stratosphere~~ using observations and ~~TOMCAT model simulations~~the TOMCAT CTM simulation, we first summarize the relationships of $O_3$ variability in this region in a shape of DAG-based on expert knowledge and literature review (as discussed and interpreted by e.g. Galytska et al., 2019; Nedoluha et al., 2015), following the procedure discussed in Sect. 3.3. Figure 3a depicts a simple linear chain from the cause $w^*$ to the outcome $O_3$ (grey nodes), with mediating variables $N_2O$ and $NO_2$ (magenta nodes). The inferred DAG, therefore, represents an effective causal structure that emerges under the influence of dynamical variability, rather than a representation of isolated chemical relationships. In particular, a positive relationship from $w^*$ to $N_2O$ (labelled A) indicates that an increase in residual vertical velocity leads to enhanced $N_2O$ concentrations. The relationship from $N_2O$ to $NO_x$ (labelled B) is negative, despite $N_2O$ being a source of $NO_x$. This apparent contradiction is an example of Simpson's paradox (Blyth, 1972) and arises because ~~upwelling~~ tropical residual velocity $w^*$ acts as a confounding ~~factor~~dynamical process, leading to an ~~anticorrelation~~ anti-correlation between $N_2O$ and $NO_2$. Namely, slower (faster) upwelling results in lower (higher) $N_2O$ concentrations and consequently longer (shorter) $N_2O$ residence time in this region, which allows more (less) time for the photochemical production of $NO_x$ from $N_2O$. ~~It is important to note that $w^*$ does not exert a direct causal influence on $NO_x$, instead, its effect is mediated through $N_2O$.~~ Consequently, higher (lower) $NO_x$ levels lead to lower (higher) $O_3$ concentrations via the $NO_x$-catalyzed ~~ozone~~ $O_3$ destruction cycle, resulting in a negative relationship ~~(labelled C)~~(labelled C, see Crutzen, 1970). Table 1 summarizes the discussed chemical-dynamical relationships in the tropical middle stratosphere as depicted in Fig. 3a.

We further justify the assumed causal DAG in Fig. 3a and validate the reliability of the causal inference method. For that, we require a benchmark dataset with known causal ground truth for validation as depicted in Fig. 3a. We consider the linear structural causal process with four time series as an example that comes from a data-generating process using the following model:

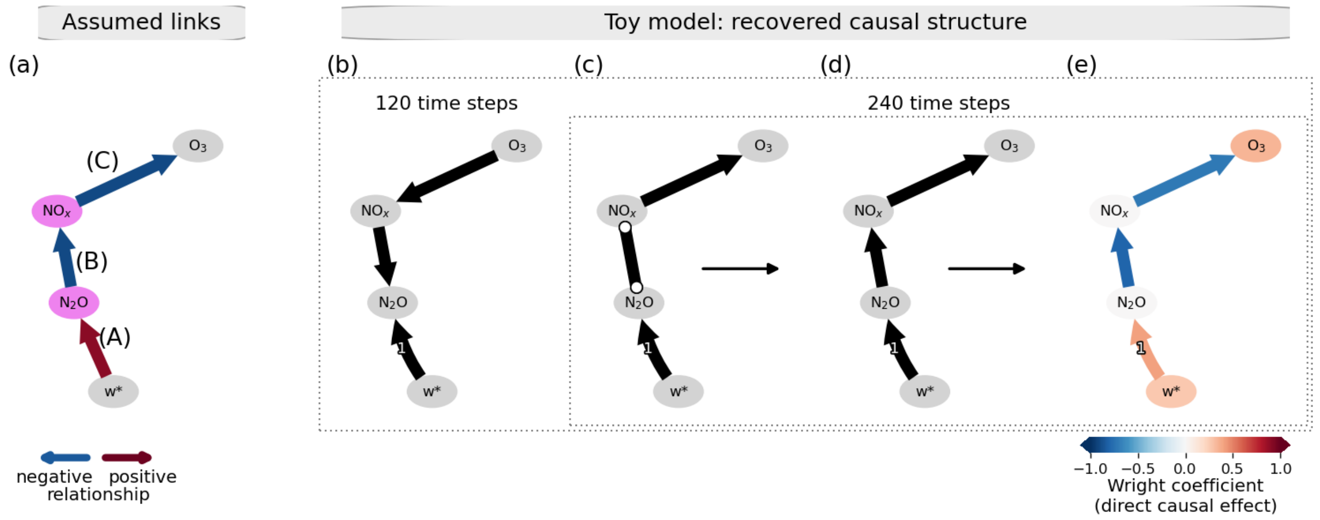

**Figure 3.** Causal justification and validation. **(a)** Assumed relationships based on expert knowledge and literature review. Magenta nodes indicate mediators in the total influence of $w^*$ on $O_3$. **(b-e)** Causal discovery applied to a generated toy model with contemporaneous (time lag = 0) and lagged (time lag > 0) dependencies based on linear partial correlation as a conditional independence test for 120 time steps (b) and 240 time steps (c), which corresponds to the DPAG. The application of the triangulated approach from Fig. 2 resulted in the DAG (d), which in turn was used as a basis for causal effect estimation, where edge colors indicate the estimated direct causal effects (e).

$$
\begin{aligned}
X_t^0 &= 0.3X_{t-1}^0 + \eta_t^0 \\
X_t^1 &= 0.3X_{t-1}^1 + 0.4X_{t-1}^0 + \eta_t^1 \\
X_t^2 &= 0.3X_{t-1}^2 - 0.94X_t^1 + \eta_t^2 \\
X_t^3 &= 0.3X_{t-1}^3 - 0.95X_t^2 + \eta_t^3
\end{aligned}
\tag{1}
$$

where $\eta_t^{\cdot}$ stands for the independent Gaussian white noise processes with variances $\sigma_{\cdot}^2$, $X^0$ depicts residual vertical velocity $w^*$, $X^1$ - the concentration of $N_2O$, $X^2$ - $NO_x$, $X^3$ - $O_3$. This set of variables and their dependencies define a toy model. Although this toy model is designed to replicate causal dependencies in the tropical middle stratosphere, it is important to emphasize that there are multiple approaches to constructing such a model. While additional variables could be further introduced based on expert knowledge and a thorough literature review, the goal of the analysis here is not to maximize the number of variables but to create an intuitive system that can simply and effectively replicate the processes under study.

Figure 3b-e illustrates the causal graphs inferred from the causal discovery algorithm (see Sect. 3.1) when applied to the toy model from Eq. (1). To evaluate the performance of the causal discovery algorithm on time series of different lengths, Fig. 3b,c show causal graphs for generated time series of 120 ~~timesteps~~ time steps (equivalent to 10 years of monthly data) and 240 ~~timesteps~~ time steps (equivalent to 20 years), respectively. When applied to the shorter time series (Fig. 3b), causal discovery fails to detect several expected connections as anticipated from Fig. 3a, likely due to limitations in the toy model, such as weaker

**Table 1.** Summary of chemical-dynamical processes in the tropical middle stratosphere.

| Label in Fig. 3a | Connection | Link type | Description |
|---|---|---|---|
| A | $w^* \rightarrow N_2O$ | positive | Transport is the primary source of stratospheric $N_2O$. In addition to determining $N_2O$ concentrations (an increase of $w^*$ leads to an increase of $N_2O$), it also affects its residence time (an increase in $w^*$ leads to a shorter residence time of $N_2O$). |
| B | $N_2O \rightarrow NO_x$ | negative | $N_2O$ is the primary source of $NO_2$ via the slow reaction $\mathbf{N_2O + O(^1D) \longrightarrow 2\,NO}$ followed by the rapid reaction $\mathbf{NO + O_3 \longrightarrow NO_2 + O_2}$. As $N_2O$ increases due to ~~a~~ faster circulation, its residence time decreases, and therefore, $NO_2$ concentrations decline (there is less time to produce $NO_2$). The resulting link is therefore negative. In the absence of dynamical variability, the relationship would appear positive, reflecting only the underlying chemical production of $NO_2$ from $N_2O$. |
| C | $NO_x \rightarrow O_3$ | negative | $NO_2$ is the primary sink of $O_3$ in the tropical middle stratosphere via the $NO_x$ catalytic cycle. |

causal signals or higher noise. In contrast, despite the plausible limitations of the toy model, most expected connections are recovered in the 20-year time series depicted in Fig. 3c. However, the inferred graph lacks directionality between $N_2O$ and $NO_2$ (shown as ○-○ edge), resulting in a DPAG. Since causal effect estimation requires a fully directed DAG, we applied the triangulation approach to resolve ambiguities, yielding the DAG in Fig. 3d. This final causal graph (Fig. 3d) serves as the basis for causal effect estimation, shown in Fig. 3e. Given approximately linear relationships among analyzed variables (see Fig. 1b-d), we assume linear causal effects and apply Wright's method (Wright, 1921). This approach is akin to linear regression slopes between two variables X and Y, with the critical distinction that the graph is used to detect and eliminate confounding influences before regression (see Sect. 3.2 and Fons et al., 2023).

### 4.2 Physical description of observed and modeled causal relations

Figure 4 presents the magnitude and sign of direct causal effects computed using Wright's approach (Wright, 1921) on causal graphs identified by the triangulation (Uleman et al., 2024) based on observations (~~top row~~a) and the TOMCAT CTM simulation (~~bottom row) over the full period~~ b) during 2004–~~2018 (panels a, d) and two subperiods: 2004–2011 (panels b, e) when $O_3$ declined, and 2012–2018 (panels c, f) when $O_3$ increased. Across all three periods,~~ 2021. The original graphs inferred by the causal discovery algorithm (see Sect. 3.1) without the application of the triangulated approach are shown in Appendix B. The causal discovery algorithm successfully identifies the anticipated connections in the observations (Fig. 4~~a–c~~a) since the signs of the direct causal effects align well with the expected processes outlined in the Introduction and as discussed in Sect. 4.1.

Notably, the positive lagged ~~links in observations in analyzed periods (panels a–c)~~ link in observations from $w^*$ to $N_2O$ ~~indicate~~ indicates that an increase in residual vertical velocity intensifies $N_2O$ transport. This enhanced transport, in turn,

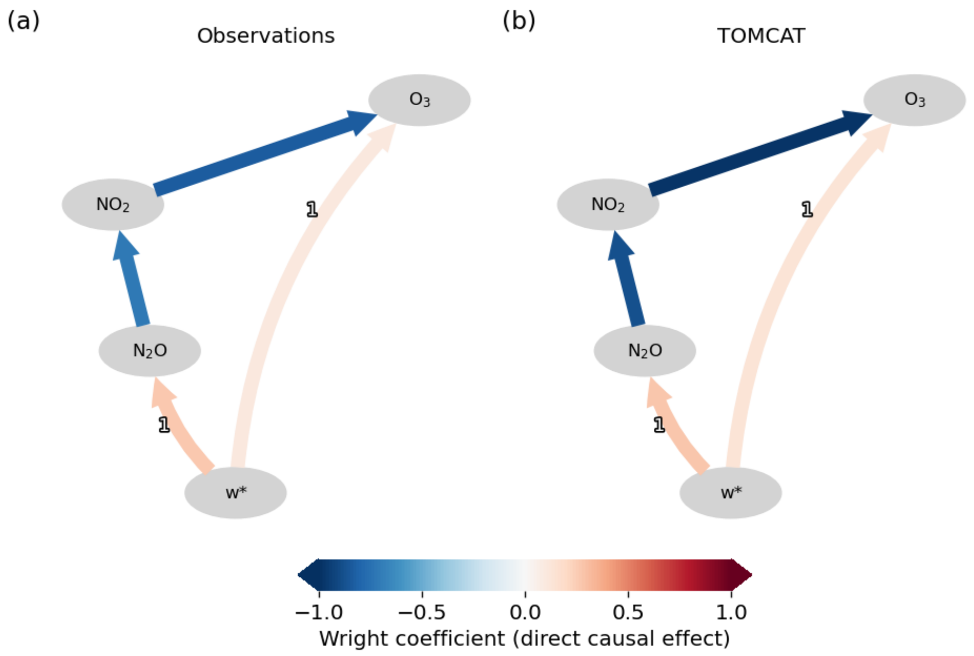

**Figure 4.** Causal inference: Magnitude and sign of the direct causal effects. The direct causal effects calculated for the observations (~~top row~~a) and the TOMCAT CTM simulation (~~bottom row~~b) from the detrended monthly anomalies for ~~the full period 2004-2018 (panels a and d), 2004-2011 (panels b and e), and 2012-2018 (panels c and f)~~2004-2021. ~~For TOMCAT, the causal graphs identified from the observations were used, and direct causal effects were then estimated with the TOMCAT data.~~ Straight arrows show the contemporaneous (time lag = 0) connections; curved arrows indicate lagged links (time lag > 0); edge color stands for the estimated direct causal effects. For TOMCAT, causal graphs identified from the observations were used, and direct causal effects were estimated using the TOMCAT data.

reduces the residence time of $N_2O$, leading to less time for $NO_2$ production via the reaction $N_2O + O(^1D) \longrightarrow NO + NO$ (also labelled B in Fig. 3a). Consequently, the causal contemporaneous link from $N_2O$ to $NO_2$ reflects the negative relationship confounded by upwelling. ~~Notably, during 2012–2018 the algorithm detected this $N_2O$ to~~ The negative contemporaneous link from $NO_2$ ~~connection at a one-month lag (and not contemporaneous at lag zero), indicating that~~ to $O_3$ indicates that lower/higher $NO_2$ ~~production occurred with a delay compared to 2004–2011 (panel b). This does not rule out the existence of a contemporaneous connection, but instead suggests that the lagged relationship was statistically more robust during that period. The emergence of this one-month lagged connection in the relationship from $N_2O$ to $NO_2$ points to a shorter $N_2O$residence time, consistent with faster transport in 2012-2018 compared to the earlier period, as further supported by~~ levels are associated with higher/lower $O_3$ concentrations, as $O_3$ loss in the tropical middle stratosphere is largely driven by catalytic $NO_x$ destruction. Causal discovery further detects a bidirected connection between $w^*$ ~~changes calculated with the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) regression model shown in Appendix ??~~and $O_3$ in the observations, indicating the presence of a latent common driver of $w^*$ and $O_3$ and that neither variable is an ancestor of the other (see Appendix B, Fig. ~~??a-c. Additional sensitivity testsconfirm the robustness of the~~ B1a). As causal effect estimation requires a DAG, we define the direction from $w^*$ to $O_3$ to quantify the strength of this link. This choice allows us to estimate the direct dynamical influence of $w^*$ on $O_3$, which is sometimes identified by a causal discovery algorithm in sensitivity tests. Based on Fig. 4, the direct influence of $w^*$ on $O_3$ is much weaker, and a mediated pathway via $N_2O$ ~~to~~ and $NO_2$ ~~lagged connection in 2012–2018 compared to the contemporaneous connection observed in 2004–2011 (see Appendix ??, Figs. ?? and ??). Additionally, the negative contemporaneous link~~ dominates. However, temperature-mediated effects could amplify the apparent strength of the connection from $NO_2$ to $O_3$~~indicates that lower/higher $NO_2$ levels result in higher/lower $O_3$ concentrations, as~~ , since enhanced upwelling induces both cooling (increasing $O_3$~~loss~~ ) and reduced $NO_x$ species. To assess this, we performed additional analysis, including temperature anomalies in the tropical middle stratosphere~~is largely driven by catalytic $NO_x$ destruction (Crutzen, 1970)~~. The inferred graph structure was not robust, likely because $w^*$ is a derived diagnostic that depends on thermodynamic fields. Removing $w^*$ altered the parent structure and prevented a direct comparison of direct causal effects shown in Fig. 4a. We therefore interpret the identified $NO_2$-mediated pathway as the dominant mechanism, while acknowledging that temperature-related effects may potentially project onto this link.

Unlike the observations, ~~the causal discovery algorithm was unable to identify the expected connections~~ in the TOMCAT CTM simulation, ~~as anticipated from the expected processes~~ the causal discovery algorithm does not fully reproduce the expected chemical-dynamical coupling as outlined in the Introduction, Sect. 4.1 and as shown in Fig. 4~~a-c. The reasons for this limitation are~~ a. In particular, the anticipated one-month lagged link from $w^*$ to $N_2O$ is not robustly detected. We found that this occurs because $O_3$ exhibits very strong contemporaneous coupling with both $N_2O$ and $NO_2$, such that conditioning on $O_3$ makes the one-month lagged $w^*$ to $N_2O$ link statistically insignificant. As further demonstrated and discussed in Appendix ~~??.~~ B, this does not indicate a lack of dynamical coupling in the TOMCAT CTM simulation. Instead, it reflects the strong shared variability among the chemical tracers as the model uses a chemically consistent scheme for all the variables. To still assess the strength of the processes represented in TOMCAT, we did not rely on the TOMCAT-derived graph. Instead, ~~we applied the graph identified from the observations and used it as a fixed structure to~~ by adopting a fixed graph structure derived from observational ground truth and expert knowledge, we can accurately estimate direct causal effects with the TOMCAT data (Fig. 4~~d-f)~~b), providing a valid and pragmatic solution for quantifying model sensitivities. Similar to observations, the direct one-month lagged $w^*$-$N_2O$ connection is estimated as significant in the TOMCAT CTM simulation for the analyzed ~~periods~~period. Additionally, $N_2O$-$NO_2$ ~~links are~~ negative contemporaneous link is slightly stronger in the TOMCAT CTM simulation (~~panels d and e~~Fig. 4b) compared to those in the observations (~~panels aand b~~Fig. 4a), with a similar causal pattern observed for the $NO_2$-$O_3$ link. ~~During 2012-2018 the strength of the~~

To ensure the robustness of the detected in Fig.4a connections in observations in the tropical middle stratosphere during the period 2004-2021, Fig. 5 demonstrates the results of the application of the causal discovery algorithm with different setups of $\tau_{max}$ (depicted on the x-axis) and $\alpha_{pc}$ (depicted with markers). $\tau_{min}$ set to zero to account for the contemporaneous connections. It should be noted that choosing a $\tau_{max}$ that is too low risks missing causal links with longer delays, which violates the assumption of causal sufficiency. However, choosing $\tau_{max}$ too high without mitigation can dilute the detection power of the causal algorithm. Specifically, a larger $\tau_{max}$ expands the search, as the algorithm tests more possible lagged pairs.

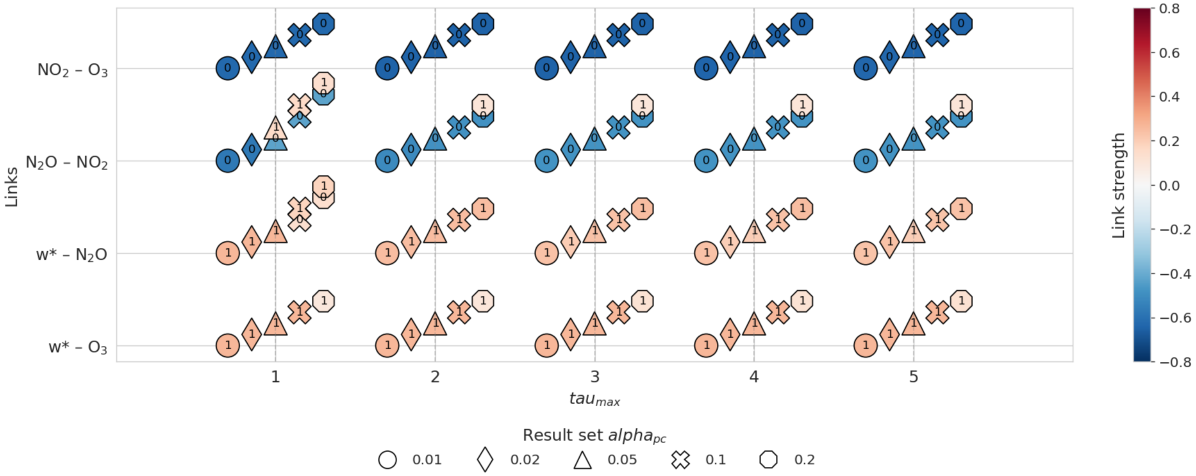

**Figure 5.** Sensitivity tests of the detected links to the choice of $\tau_{max}$ (x-axis) and $\alpha_{pc}$ (depicted with markers) for the observations for the period 2004-2021. For all tests $\tau_{min}$ = 0. Only link pairs shown in Fig. 4 are considered.

This leads to larger conditioning sets, which can further reduce the effect size and detection power. Therefore, it is important to condition only on a few relevant variables that actually explain the relationship (Runge et al., 2019). Sensitivity tests from Fig. 5 show very similar results for different configurations of $\tau_{max}$ and $\alpha_{pc}$. The chemical connections related to $N_2O$-$NO_2$ and $NO_2$-$O_3$ pairs are robustly detected as contemporaneous across all experiments. The dynamical connections $w^*$-$N_2O$ and $w^*$-$O_3$ are robustly detected with a one-month lag.

A further analysis of the sensitivity experiments reveals an additional feature in the $N_2O$-$NO_2$ relationship. A positive one-month lagged link from $N_2O$ to $NO_2$ ~~from the~~ is detected across all tested $\tau_{max}$, but primarily at relaxed significance thresholds ($\alpha_{pc}$=0.2). For $\tau_{max}$=1, the link is also identified at $\alpha_{pc}$=0.1 and 0.05. Such a positive lagged connection is physically plausible, as $NO_2$ is produced from $N_2O$, as discussed in Table 1, and a delayed response may emerge at the monthly timeseries. However, given its sensitivity to the choice of $\alpha_{pc}$, this link cannot be considered a robust pathway.

### 4.3 Process-oriented analysis

We adopt the process-oriented causal analysis (Nowack et al., 2020; Karmouche et al., 2023; Eyring et al., 2024; Debeire et al., 2025) to major drivers of tropical middle stratospheric $O_3$ variability to further understand the robustness of the connections during different regimes. Given that $N_2O$, $NO_2$, and $O_3$ exhibit a strong QBO signal (Chipperfield et al., 1994; Tian et al., 2006; Park et al., 2017; I
, we mask the data to easterly and westerly shear zones (see Sect. 3.4). This approach makes it possible to explore the regime-dependent robustness of causal relationships. A robust connection between specific variables indicates that the particular link is consistently detected across multiple resampled datasets. A robust connection also suggests that the relationship is less sensitive to variations in the data, providing higher confidence that the detected connection is not a result of random fluctuations or sampling variability. It is also important to note that the analyzed period includes an unprecedented QBO disruption in 2016

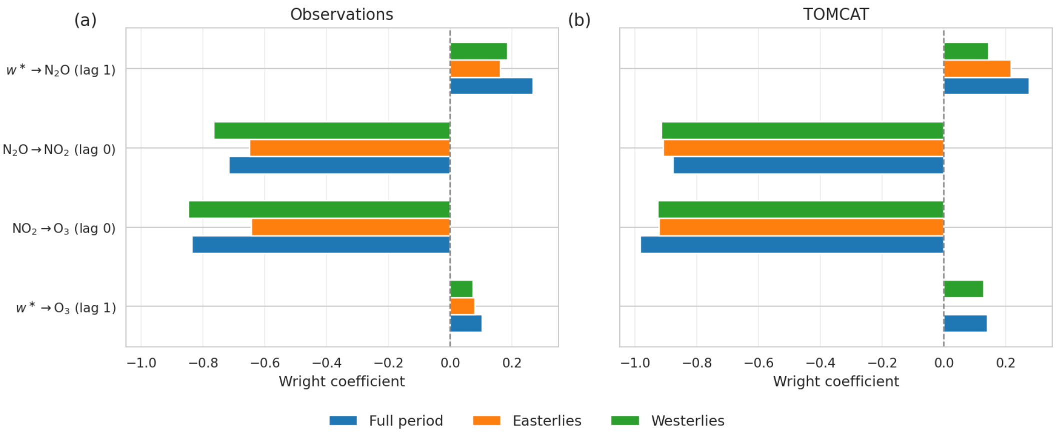

**Figure 6.** Regime-oriented direct causal effects from observations (a) and the TOMCAT CTM simulation (b) for the full 2004-2021 period (blue), and for easterly (orange) and westerly (green) QBO regimes.

(Tweedy et al., 2017; Match and Fueglistaler, 2021), which stalled the descent of the easterly shear toward 10 hPa and caused anomalous upwelling and wind patterns, temporarily altering transport and mixing in the tropical middle stratosphere. To assess how QBO phase affects the strength of the identified relationships, we keep the causal graph inferred from the full period and re-estimate the link strengths separately for easterly and westerly QBO conditions. This approach isolates regime-oriented changes in coupling strength without altering the underlying network structure. Figure 6 illustrates the estimated direct causal effects for both observations (panel a) and the TOMCAT CTM simulation (panel ~~f)~~ b) for the full period 2004-2021 (blue), which also corresponds to direct causal effects shown in Fig. 4a,b, easterly (orange), and westerly phase of the QBO.

The dynamical positive link from $w^*$ to $N_2O$ at a lag of one month shows slightly reduced magnitude in observations when separating into QBO phases compared to the full period. In the TOMCAT CTM simulation, the link strength is similar to ~~the observational one (panel c) , highlighting the agreement between different data sources~~observations and also slightly reduces across QBO phases. The negative contemporaneous chemical coupling between $N_2O$ and $NO_2$ is stronger in the TOMCAT CTM simulation compared to observations and of similar strength across phases. In observations, the absolute magnitude is slightly larger during the westerly phase than during the easterly phase. The contemporaneous negative link from $NO_2$ to $O_3$ is likewise stronger in the TOMCAT CTM simulation compared to the observations. In the observations, this link weakens during the easterly QBO phase, whereas in the TOMCAT CTM simulation its magnitude is comparatively stable across QBO phases.

The imposed one-month lagged connection from $w^*$ to $O_3$ is the weakest among the analyzed pathways and substantially smaller than the chemically mediated pathway. In the TOMCAT CTM simulation, no link is detected for the easterly QBO phase. This phase-dependent absence of this connection points to its limited robustness. This also indicates that the direct influence of the residual circulation on $O_3$ variability is predominantly mediated through $N_2O$ and $NO_2$, rather than through a direct dynamical effect. Overall, the sign of all connections is robust across datasets and QBO phases. In the observations, the chemical links (from $N_2O$ to $NO_2$ and from $NO_2$ to $O_3$) tend to strengthen during the westerly QBO phase.

### 4.4 Causal effects across different time lags

While the causal discovery algorithm, along with causal effects estimation of the direct links, offers a general overview of whether the methodology captures the expected dependencies, one can further study the propagation of causal effects through the causal graph across different time lags. The total causal effects (for further discussions, see Appendix C), which are not necessarily just depicted by a single arrow in the causal graph, can be derived from direct causal effects using Wright's path analysis for specific relationships across different time lags. Figure 7 shows total ~~and direct~~ (lines) and direct (labels) causal effects across different time lags for a selection of different (X, Y) pairs of variables, where a positive connection, similar to Fig. 4, indicates that an increase in X leads to an increase in Y after a time lag ($\tau$). Due to the limited number of variables in the causal graphs, we do not analyze the role of mediators for specific connections, as their influence is straightforward. For example, the total effect from $w^*$ to $NO_2$ is mediated solely by $N_2O$, as shown in Fig. 4. However, for the analyses that involve more complex causal graphs, similar to Fons et al. (2023), we recommend estimating the contribution of various mediators on a specific set of connections.

~~The total and direct~~ Figure 7 demonstrates that the total causal effects across different time lags in the TOMCAT CTM simulation (dashed lines) closely align with observations ~~across analyzed periods~~(solid lines) across analyzed regimes, indicating that the model reproduces not only the sign but also the temporal structure of the effects. However, the ~~causal effects~~ amplitudes in the TOMCAT CTM simulation are slightly ~~stronger than those in observations, particularly for the effect of~~ larger, suggesting a somewhat stronger coupling between analyzed variables in the simulation. The direct effect of $w^*$ ~~on other variables (see dashed lines in Fig. 7a-c). The direct causal effect of residual vertical velocity $w^*$~~ on ~~the transport of~~ $N_2O$ ~~into the stratosphere~~ (Fig. 7a) ~~reveals a peak in the positive connection~~ exhibits a clear maximum at a lag of ~~four months in the observations and the TOMCAT CTM simulation across all three periods. The total effect~~ around three months for the full period 2004–2021 and during the easterly QBO phase. This peak defines the characteristic adjustment timescale of dynamical transport. The total effects of $w^*$ on $NO_2$ (Fig. 7b) and on $O_3$ (Fig. 7c and Appendix C) ~~, both clearly mediated by~~ also reach their maxima at approximately three months, consistent with a sequential mediation through $N_2O$ ~~, reaches its maximum at a lag of approximately 3–4 months in both cases. This indicates the timeframe within which $w^*$ must act to influence these chemical constituents. Observations and the TOMCAT CTM simulation agree that the total effect of $w^*$ on~~ and subsequently $NO_2$~~and $O_3$ during the 2012–2018 period (Fig. 7b,c) is delayed and significantly weaker than in other periods. This reduced total effect from~~. We can further decompose the total effect of $w^*$ on ~~$NO_2$ and~~ $O_3$ ~~indicates weaker connections between the variables, potentially due to factors such as a shorter residence time of $N_2O$, leading to a lower $NO_2$ production and , as a result, O~~into direct and indirect contributions. As shown by Fig. 4a and Fig. 6a, the direct one-month lagged $w^*$ on the $O_3$ ~~increase (shown in Fig. 1a and consistent with changes from Fig. ??).~~

~~The $N_2O$ on $NO_2$ relationship (Fig. 7d) shows close alignment for the subperiods 2004-2011 and 2012-2018, particularly after a lag of two-three months, with~~ pathway is approximately 0.10 in the observations and 0.14 in the TOMCAT CTM simulation~~detecting slightly weaker relationships. This suggests no significant differences in the influence of~~. The indirect contribution, mediated via $N_2O$ ~~on~~ and subsequently $NO_2$~~after a lag of two-three months in both subperiods. However, the~~

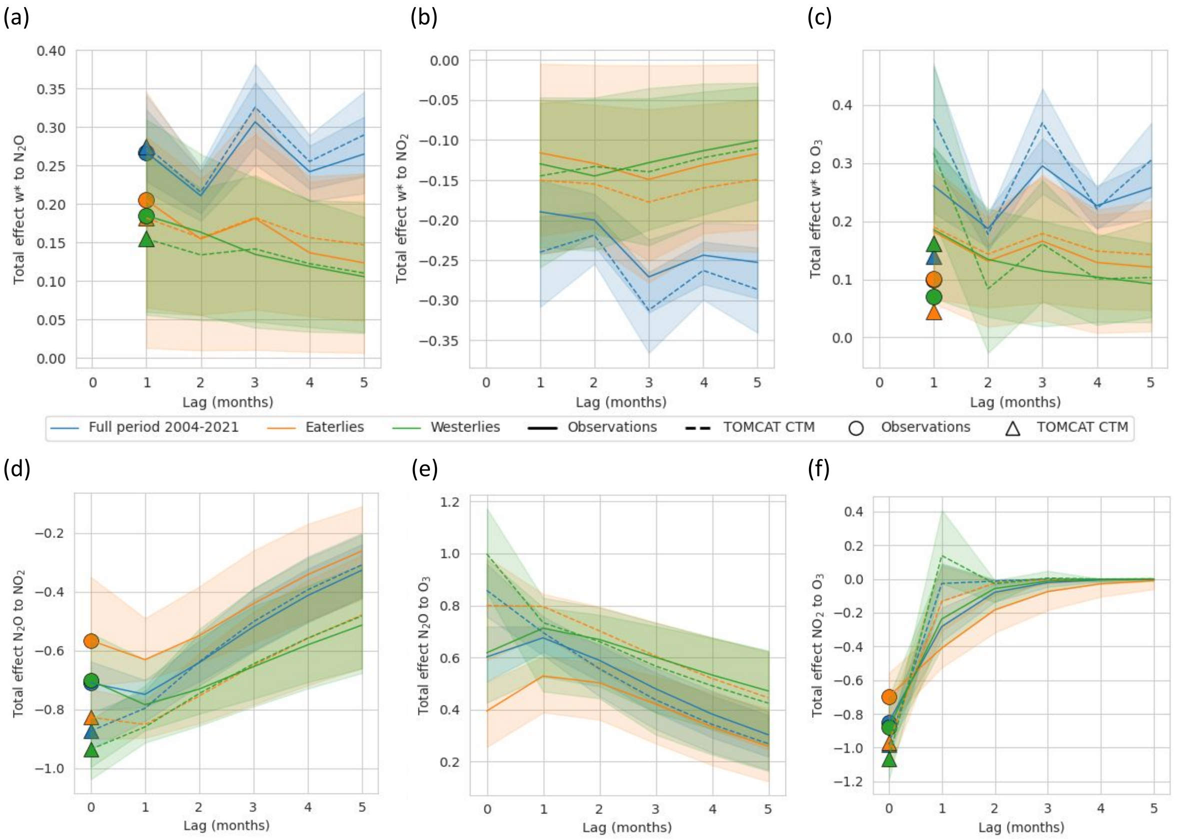

**Figure 7.** Total ~~and direct~~ causal effects across different time lags ~~(~~in months ~~)~~ from observations (solid lines) and the TOMCAT CTM simulation (dashed) for (a) $w^*$ on $N_2O$, (b) $w^*$ on $NO_2$, (c) $w^*$ on $O_3$, (d) $N_2O$ on $NO_2$, (e) $N_2O$ on $O_3$, (f) $NO_2$ on $O_3$. All plots show the results obtained for ~~2004-2018~~ 2004-2021 (blue), ~~2004-2011~~ easterly (orange), and ~~2012-2018~~ westerly QBO phase (green). The markers indicate direct causal impact presented in Fig. 4 and 6. The shading corresponds to the 90% bootstrap confidence interval.~~In panel (f), the blue and green curves largely overlap, and therefore only the blue line is visible.~~

~~largest differences are observed at lags prior to two months, where the more significant connection for the period 2012-2018 first appears at lag one and is stronger between lags one and two compared to the 2004-2011 period. This pattern is related to the $N_2O$-$O_3$ causal relationship (Fig. 7e)~~, ~~where the total influence of $N_2O$~~ reaches 0.16 in the observations and 0.23 in the simulation (not shown here). Therefore, the total effect of $w^*$ on $O_3$ ~~is delayed by one month.~~

~~The $NO_2$ on $O_3$ relationship (~~shown in Fig. 7~~f) peaks at lag zero in both observations and the TOMCAT CTM simulation, consistent with fast chemical reactions. Notably, this is the only connection where the effect decays rapidly, reaching near-zero values at~~ c at a lag of one month ~~for the full period 2004-2018 and 2012-2018 and around three months for the 2004-2011 period. The longer persistence of the causal effect of $NO_2$ on $O_3$ provides direct evidence of extended exposure of $O_3$ to the $NO_x$ catalytic cycle, contributing to the ozone decline observed during 2004-2011 (also shown in Fig. 1).~~

~~The specific causal relationships across different time lags provide insights into the observed changes in the analyzed species (Appendix D further discusses lagged dependencies of causal effects). The changes in analyzed variables were also investigated using the LOTUS regression model, with a discussion provided in Appendix **??**. By comparing and combining the causal inference approach (including causal effects across different time lags) with the LOTUS regression model, we link the one-month delayed influence of $w^*$ on $O_3$ through~~ therefore represents the sum of both pathways, yielding approximately 0.26 for the observations and 0.37 for the TOMCAT CTM simulation over the full analyzed period. The same analysis is performed for the easterly and westerly QBO phases, but an analogous description is not discussed further. Conditioning on both $N_2O$ and $NO_2$ ~~mediators during the period 2012-2018 to the observed increase in $O_3$ levels (see Appendix **??**, Fig. **??**). This increase is a consequence of the delayed $NO_x$ production from N~~does not alter the magnitude of the mediated effect, indicating that the dominant indirect pathway proceeds sequentially through $w^*$, $N_2O$, ~~which is consistent with the negative trend in NO~~$NO_2$~~(Fig. **??**h). In contrast, during 2004-2011, the $N_2O$-$NO_2$ relationship at lag zero was stronger, leading to a higher production of $NO_x$ species at lag zero compared to 2012-2018, which explains the positive trend in $NO_2$ during 2004-2011 (Fig. **??**g).~~

~~Process-oriented causal effect estimation~~

~~We adopt the process-oriented causal analysis (Nowack et al., 2020; Karmouche et al., 2023; Eyring et al., 2024; Debeire et al., 2025) to major drivers of tropical middle stratospheric~~ , and $O_3$~~variability to further understand the robustness of the connections during various regimes. Given that~~ .

The $N_2O$ ~~,~~ on $NO_2$ ~~, and $O_3$ exhibit a strong QBO signal (Chipperfield et al., 1994; Tian et al., 2006; Park et al., 2017), apart from splitting the analyzed time period to 2004-2011 and 2012-2018, we also mask the data to westerly and easterly shear zones in the tropical middle stratosphere (see Sect. 3.4) . This approach makes it possible to explore the regime-dependent and period-specific robustness of causal relationships. A robust connection between specific variables indicates that the particular connection is consistently detected across multiple resampled datasets. A robust connection also suggests that the relationship is less sensitive to variations in the data, providing higher confidence that the detected connection is not a result of random fluctuations or sampling variability. It is also important to note that the 2012-2018 period includes an unprecedented QBO disruption in 2016 (Tweedy et al., 2017; Match and Fueglistaler, 2021), which stalled the descent of the easterly shear toward 10 hPa and caused anomalous upwelling and wind patterns, temporarily altering transport and mixing in the tropical middle stratosphere.~~

~~Regime-oriented direct causal effects from observations for the complete 2004-2018 period (middle panel) and corresponding regimes. Each graph is labelled with the total number of data points (months) characterized by each regime. Similar to Fig. 4, the color of the arrows indicates the value of the direct causal effect. All nodes shown in grey were hypothesized to be auto-dependent with a lag of one.~~

~~Figure **??** summarizes the causal graphs derived from the observations. The central causal graphs resemble those from Fig. 4a-c. To refine the process-oriented analysis, we further masked the data according to easterly and westerly wind shear regimes within the 2004–2011 (top) and 2012–2018 (bottom row) periods. It should be noted that the shorter subperiods may limit the robustness of causal discovery, and the corresponding sample sizes are therefore indicated in brackets for each regime.~~

~~For instance, during easterly wind shear in 2012–2018, only 19 months of data were available, preventing the detection of meaningful causal connections.~~

~~Figure ?? shows a positive one-month lagged connection from $w^*$ to N~~relationship (Fig. 7d) strengthens gradually with increasing lag, with close agreement between observations and the TOMCAT CTM simulation after two to three months. Differences are largest at short lags, where the observational estimate shows stronger variability. The observations suggest that this link appears weaker during easterly QBO shear, linked to vertical ascent due to the strengthening of tropical upwelling (Baldwin et al., 2001). The total $N_2$~~O that emerges consistently~~ O to $O_3$ effect (Fig. 7e) increases from lag zero to lag one (in the full ~~2004–2018 record and under both easterly and westerly shear regimes for this longer period. In the shorter subperiods, when masking data for the wind shear, this connection emerges during westerly wind shear in 2012-2018. This aligns with the understanding that westerly winds are linked to vertical descent due to the weakening of tropical upwelling (Baldwin et al., 2001). During the regime of the easterly wind shear, the causal discovery did not detect the connection from $w^*$ to $N_2O$ for the 2004–2011 and 2012–2018 periods, which is related to the very limited sample size. Further consistently detected connections across nearly all analyzed subperiods are the negative contemporaneous relationship from $N_2O$ to~~ period) and then gradually decreases toward longer lags. Here, observations also show a weaker total effect during easterly QBO phase. These results are also consistent with direct causal effects analyzed for different QBO regimes (see Fig. 6). The $NO_2$ ~~, and from $NO_2$ to~~ on $O_3$ ~~. This aligns with the connections shown in Fig. 4a-c, highlighting the strong dependence and robustness of this chemical relationship across different regimes. Figure ?? further reveals regime-dependent differences in the causal links, particularly in the chemical connection from N~~effect (Fig. 7f) peaks at lag zero and rapidly decreases, approaching zero after about one month. This behavior is consistent with fast $NO_x$-driven $O_3$ chemistry. A slightly longer persistence during the earlier subperiod suggests a more sustained influence of $NO_2$ ~~O to $NO_2$. This link appears weaker during westerly shear, associated with reduced upwelling, and stronger during easterly shear, reflecting enhanced upwelling. Although the relatively short subperiods analyzed may influence the robustness of these results, Fig. ?? illustrates how causal discovery methods can capture both regime-specific and period-dependent interactions in stratospheric chemistry.~~ on $O_3$ during that time.

## 5 Conclusions

This study applies causal inference to ~~better understand~~ quantify the contributions of chemical-dynamical drivers ~~to~~ that control $O_3$ variability in the tropical middle stratosphere. ~~By using~~ Using a causal discovery algorithm ~~on observational data, we identify distinct causal relationships~~ applied to observations over the 2004–~~2018 period, as well as within two key subperiods: 2004~~2021 period, we robustly identify a dominant chemical–~~2011 and 2012-2018. The selection of these subperiods is motivated by contrasting O~~dynamical pathway, in which variability in residual vertical velocity $w^*$ modulates $N_2O$, subsequently affecting $NO_2$ and ultimately $O_3$~~changes, namely declining levels in the first subperiod and increasing levels in the second~~. The resulting causal graphs ~~show the capability~~ are then used within the causal inference framework to quantify the temporal contribution of specific variables ~~, namely $w^*$, $N_2O$, and $NO_2$~~ on monthly $O_3$ variability ~~. Further, we used~~ under different QBO phases and to separate the direct and mediated effects. In the TOMCAT CTM simulation, strong shared variability among the chemical tracers limited the ability of the discovery algorithm to detect expected dynamical coupling. Therefore, we applied observational graphs based on triangulation (Uleman et al., 2024) to estimate causal relationships in the TOMCAT monthly data. This example illustrates a practical workaround for cases where models or datasets do not yield the expected causal structure through discovery. Importantly, estimating direct or total causal effects does not require that the graph be learned by an algorithm since a graph ~~based on~~ informed by expert knowledge and supported by the literature can serve as a valid alternative (Fons et al., 2023). In our case, the close agreement between the observationally derived graph and the main relationships anticipated from established chemical-dynamical interactions (see Fig. 4 and as discussed in the Introduction~~and shown in Fig. 1~~) demonstrates that the discovery algorithm can indeed recover the expected structure when applied to suitable data~~, highlighting its potential for uncovering causal connections even when they are not known a priori.~~ . Additionally, the toy model validation further demonstrates reliability under finite-sample conditions (Sect. 4.1). Therefore, the methodology adopted in this study, which integrates triangulation (Uleman et al., 2024) for the construction of causal graphs and an algorithm for causal effect estimation, demonstrates a comprehensive approach to causal inference. This ensures the robustness of the analyzed system and facilitates the quantification of specific connections in a physically meaningful domain.

~~In particular, the estimated direct and total causal connections yielded similar results for~~ Direct causal effects applied to the observations and the TOMCAT CTM simulation ~~. However, the connections involving residual vertical velocity $w^*$ were somewhat stronger in the TOMCAT CTM simulation than in the observational data, especially beyond a lag of approximately one month. We found that a one-month lagged connection from $N_2O$to~~ reveal that tropical middle-stratospheric $O_3$ variability is dominated by an indirect $NO_2$~~during the 2012~~-mediated pathway, consistent with previous studies of chemical~~–2018 period robustly contrasted with the contemporaneous link observed during 2004–2011 and 2004-2018. This suggests that $NO_2$ production during 2012–2018 took longer, likely due to faster transport of $N_2O$ and , consequently, its shorter residence time. Supporting this interpretation, the total causal effect across different time lags~~ dynamical coupling (see Fig. 4 and e.g. Portmann et al., 2012 . This mechanism of identified connections captures variability under different QBO phases via regime-oriented analysis and is also supported by sensitivity tests. For example, the direct influence of $w^*$ on ~~$NO_2$, as well as $w^*$ on~~ $O_3$ ~~(both mediated by $N_2O$) shows a one month lagged influence during 2012–2018 compared to 2004–2011. This lagged influence is consequently seen in the direct effect of~~ is weak and not robust across QBO phases. The total causal effects that consist of direct and mediated pathways peak at a lag of approximately two-three months (as discussed in Sect. 4.4, Fig. 7), indicating a propagation of the dynamical signal from $w^*$ through $N_2O$ ~~on~~ and $NO_2$ ~~and the total effect of $N_2O$on O~~to $O_3$. ~~These differences in $w^*$-$N_2O$-$NO_2$ connections for the analyzed subperiods resulted in reduced $NO_x$ formation during 2012-2018 and, consequently, lower catalytic $O_3$ destruction. It is important to emphasize that a deeper understanding of the physical mechanisms underlying the detected causal relationships is essential, especially when considering different dynamical regimes or processes. In this study, causal inference was applied separately to easterly and westerly wind shear regimes, revealing that the causal link from~~ The process-oriented analysis for different QBO phases applied to the observations suggests that the chemical links between $N_2O$ ~~to~~ and $NO_2$~~was stronger during easterly shear than during westerly shear .~~, and $NO_2$ and $O_3$ strengthen during westerly shear compared to easterly shear (see Fig. 6). Additional sensitivity tests, including temperature anomalies in the tropical middle stratosphere, did not yield a robust and interpretable graph. Temperature-related effects may therefore partly project onto the identified $NO_2$-mediated pathway, but within the present framework their contribution cannot be robustly separated and quantified. ~~However, the relatively short subperiods analyzed may affect the robustness of these findings and should be considered when interpreting the results.~~

Our study highlights the pivotal role causal inference can play in identifying, disentangling, and quantifying complex physical and chemical-dynamical processes in the stratosphere. This work lays the foundation for extending the application of causal inference to other areas ~~of stratospheric chemistry and stratospheric~~ involving complex chemical-dynamical interactions. Given its potential, this approach is particularly valuable for systems with connections that are not ~~yet fully understood. Such an~~ well understood. An integration of causal reasoning into data-driven science ~~enhances~~ will help us to enhance our understanding of ~~processes and supports~~ these complex processes and will support the development of robust methodologies that combine machine learning with statistical approaches. This integration is especially relevant for Earth and environmental sciences, as it benefits both observational and modeling studies. Causal inference has already proven to be an effective tool for climate model evaluation, particularly by enabling comparisons between causal graphs derived from models and those based on observations. This emerging methodology is gaining traction in process-oriented climate model evaluation (Debeire et al., 2025; Karmouche et al., 2023; Galytska et al., 2023; Nowack et al., 2020) and offers valuable insights into the physical mechanisms driving the varying performance of different models. Notably, causal model evaluation not only identifies physically based connections but can also reveal the processes that are poorly represented in models. We also identify a key direction for future research focused on cross-evaluating the process-oriented performance of different CTMs and coupled chemistry-climate models (CCMs). Such efforts could provide valuable insights into model accuracy and reliability of CCMs, ensuring that the distribution of simulated species or processes is not driven by incorrect or unknown factors. Additionally, integrating causal inference with well-established statistical methods could further advance stratospheric studies, particularly in analyzing the variability of chemical species. Testing these approaches across different chemical and dynamical processes in various stratospheric regions represents a promising avenue for improving model evaluation and understanding complex interactions.

*Code and data availability.* The code used to reproduce results, including figures for this manuscript, will be accessible in Zenodo and in the following GitHub repository https://github.com/EyringMLClimateGroup/. The causal discovery algorithm is implemented in the Python package Tigramite is released under GNU General Public License v3.0. Tigramite v5 is publicly available on Zenodo https://doi.org/10.5281/zenodo.6247837 Runge (2022) or via https://github.com/jakobrunge/tigramite, last access: 01.08.2025.

Transformed Eulerian mean data from the ERA5 reanalysis (monthly means) is available on Zenodo (https://zenodo.org/records/7081721, Serva, 2022). MLS $N_2O$ v5.01 Lambert et al. (2020) is publicly available https://disc.gsfc.nasa.gov/ upon registration. OSIRIS $O_3$ and $NO_2$ data are available at https://research-groups.usask.ca/osiris/data-products.php#. QBO equatorial winds are provided by the Institute of Meteorology and Climate Research at the Karlsruhe Institute of Technology (KIT) and are publicly available on Zenodo (https://zenodo.org/records/18850668, Kerzenmacher and Braesicke, 2026). Data from the TOMCAT CTM simulation is available upon request from the authors.

## Appendix A: ~~Stratospheric trends based on LOTUS regression model~~

~~To review the latitude–altitude changes in the distribution of analyzed species, we applied the open-source regression model Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS, v0.8.3) (Godin-Beekmann et al., 2022). LOTUS employs the classical multiple linear regression method to estimate time series variability from proxies using the general least squares approach. In this study, we used the following proxies: QBO, ENSO, the 11-year solar cycle, stratospheric aerosol optical depth (sAOD), and a long-term trend. Independent linear trend (ILT) terms were introduced to assess changes in the analyzed species during 2004–2011 and 2012–2018. All data were monthly mean time series, deseasonalized within the LOTUS model using Fourier components to represent annual and semi-annual variations. The fitting of the deseasonalized time series followed the approach described by Godin-Beekmann et al. (2022), with the exception that QBO equatorial wind data from KIT was used and standardized at three vertical levels: 10, 30, and 50 hPa. Although the LOTUS model does not show statistically significant changes at the 90% confidence level for some species in certain subperiods, these intervals are retained to better understand the processes driving $O_3$ variability.~~

~~Latitude–altitude distribution of changes in (a-b) $w^*$ in mm/s, (d-e) $N_2O$, (g-h) $NO_2$, and (j-k) $O_3$ in % from the LOTUS regression model for two subperiods 2004-2011 and 2012-2018 and their corresponding vertical profiles in the tropical stratosphere in panels c,f,i,l. Hatched areas show changes that are not statistically significant at 90% confidence. The dashed boxes indicate the region of the tropical middle stratosphere.~~

## Appendix A: ~~Sensitivity tests~~

~~Choosing a $\tau_{max}$ that is too low risks missing causal links with longer delays, which violates the assumption of causal sufficiency. However, choosing $\tau_{max}$ too high without mitigation can dilute the detection power of the causal algorithm. Namely, a larger $\tau_{max}$ expands the search, as the algorithm tests more possible lagged pairs. This leads to larger conditioning sets, which can further reduce the effect size and detection power. Therefore, it is important to condition only on a few relevant variables that actually explain the relationship (Runge et al., 2019). Figures **??** and **??** demonstrate the results of the application of the causal discovery algorithm for observational datasets for the period 2004-2011 and 2012-2018 correspondingly with different setups of $\tau_{max}$ (depicted on the x-axis) and $\alpha_{pc}$ (depicted with markers). $\tau_{min}$ set to 0 to account for the contemporaneous connections. Sensitivity tests show very similar results based on different setups of $\tau_{max}$ and $\alpha_{pc}$. During the 2011-2018 period, the link from $N_2O$ to $NO_2$ is robustly detected as one month lagged, even with $\tau_{max}$ set to > 1 (**??**). The chemical connections from $N_2O$ to $NO_2$ and from $NO_2$ to $O_3$ are robustly detected as contemporaneous across all experiments.~~

~~Sensitivity tests for the observed connections for the period 2004-2011 based on the application of the LPCMCI algorithm with different $\tau_{max}$ (1, 2, 3, 5, and 10) plotted on the x-axis and different $\alpha_{pc}$ (i.e. 0.01, 0.02, 0.05, 0.1, 0.2) depicted with markers. For all tests $\tau_{min}$ = 0. Only the links discussed in Sect. 4.1 are shown.~~

~~Same as Fig. **??** but for the period 2012-2018.~~

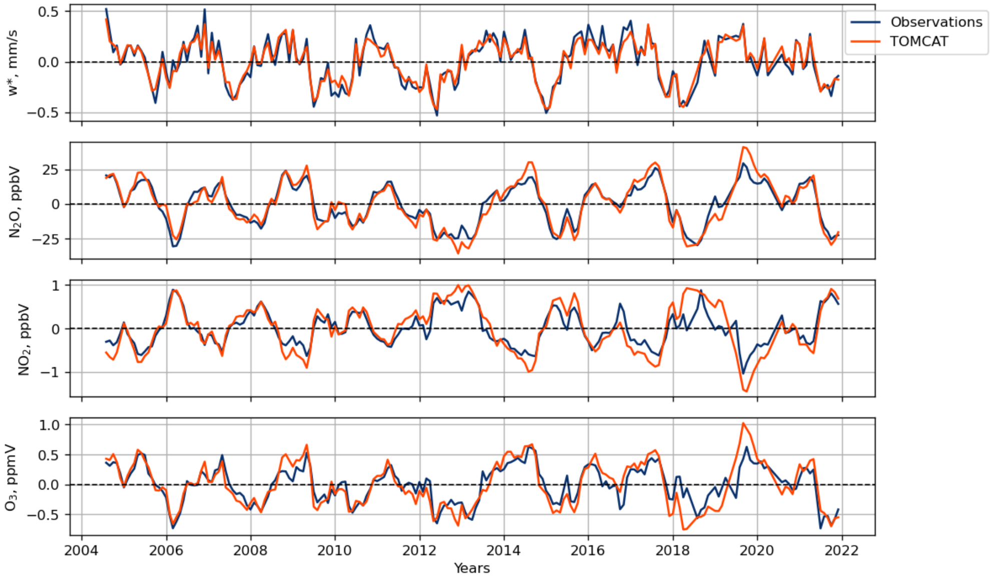

**Figure A1.** Monthly mean detrended anomalies over tropical middle stratosphere during ~~2004-2018~~ 2004-2021 in observations (blue) and the TOMCAT CTM simulation (orange). The TOMCAT CTM data is masked to match the occurrence of the observations.

## Appendix A: Comparison of observations and the TOMCAT CTM simulation

The detrended monthly mean anomalies ~~over~~ in the tropical middle stratosphere from observations and the TOMCAT CTM simulation that were used for the causal inference are shown in Fig. A1. Monthly anomalies from the TOMCAT CTM simulation closely align with observations.

Figure A2 depicts lagged dependencies from observations (blue) and the TOMCAT CTM simulation (orange) in the tropical middle stratosphere during ~~2004-2018~~ 2004-2021 using the RobustParCorr class, i.e., computing lagged correlations after transforming all marginal distributions to Gaussians. Overall, the model reproduces both the sign and the magnitude of the observed relationships across most variable pairs and lags, with correlations generally decreasing in magnitude with increasing time lag. The autocorrelations of the analyzed variables are mostly similar between observations and the TOMCAT CTM simulation, except for ~~N~~NO$_2$~~O~~, which exhibits slightly ~~weaker~~ stronger autocorrelation in the TOMCAT CTM simulation across all time lags compared to observations. The ~~connection~~ connections between $w^*$ and O$_3$ (first row, last column), N$_2$O and NO$_2$ ~~and $w^*$ and~~ (second row, third column), and O$_3$ ~~(top row, last two columns) is slightly stronger~~ and NO$_2$ (last row, third column) also show stronger lagged dependencies in the TOMCAT CTM simulation in comparison to observations.

Figure A3 depicts kernel density estimates of the joint and marginal (diagonal panels) densities of $w^*$, N$_2$O, NO$_2$, and O$_3$ in the tropical middle stratosphere. Color coding corresponds to Fig. A2. The contours illustrate the covariance structure between the variables, and the annotated $\tau$ values indicate the lag at which the strongest association is found. The overall orientation of

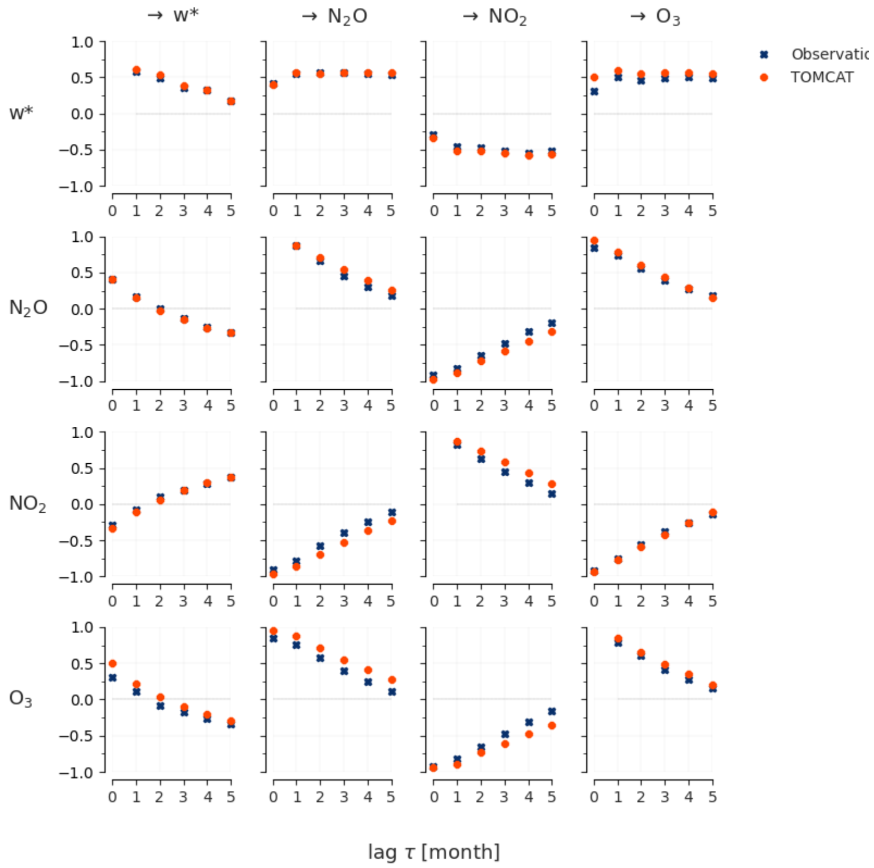

**Figure A2.** Lagged dependencies ~~over~~ in the tropical middle stratosphere during ~~2004-2018~~ 2004-2021 in observations (blue) and the TOMCAT CTM simulation (orange) based on the RobustParCorr class.

the contours is similar between observations and the TOMCAT CTM simulation, suggesting that the model captures the main sign of the relationships.

~~To further explore potential challenges in detecting anticipated causal graphs from the TOMCAT CTM simulation, we performed a set of sensitivity tests. Figure **??** illustrates the successful detection of anticipated connections ($w^*$-$N_2O$-$NO_2$-$O_3$ chain) in observations (panel~~ It is important to highlight that the lagged dependencies (Fig. A2) and density plots (Fig. A3) only quantify pairwise co-variability. They do not separate direct from indirect effects and do not condition on other variables. Therefore, the lag at which the correlation is strongest should not be interpreted as the lag of a direct causal interaction. In a system with common dynamical forcing and persistence, pairwise correlations can reflect mixed pathways operating on different time scales.

## Appendix B: Graphs inferred by the causal discovery algorithm and sensitivity testing

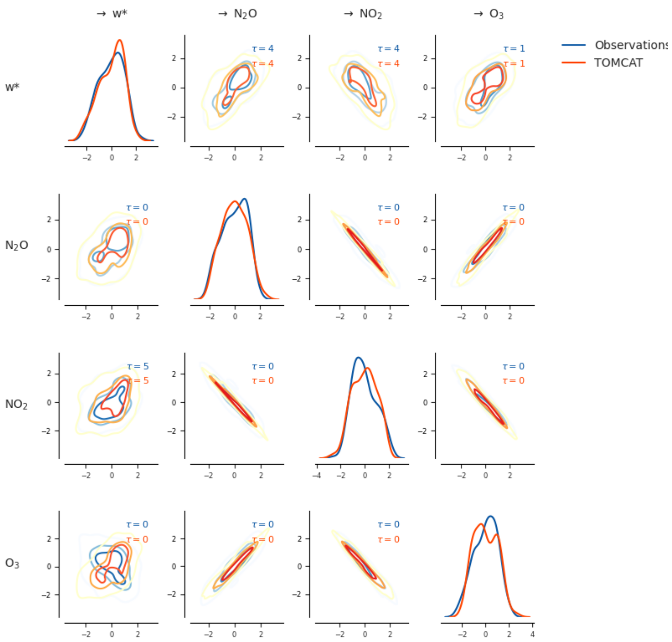

**Figure A3.** Density estimates of the joint and marginal densities over tropical middle stratosphere during ~~2004-2018~~ 2004-2021 in observations (blue) and the TOMCAT CTM simulation (orange) based on the RobustParCorr class.

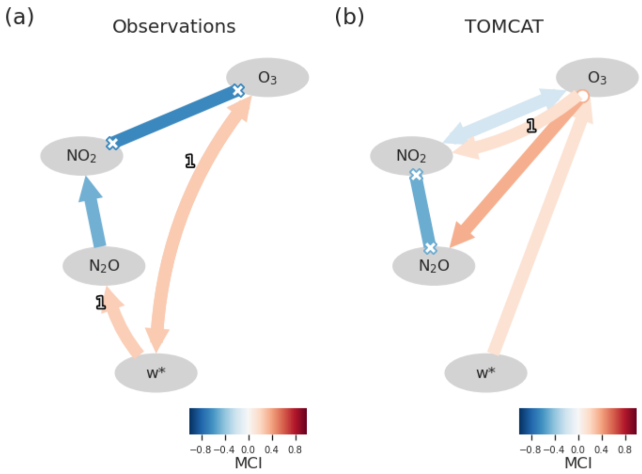

**Figure B1.** Causal graphs detected by the causal discovery algorithm from (a) observations (b) the TOMCAT CTM simulation for the period 2004-2021 with significance level $\alpha_{pc}$=0.05 and $\tau_{max}$=2.

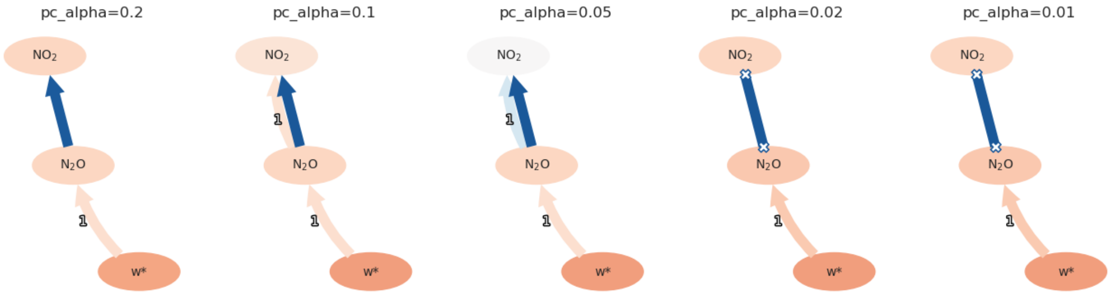

**Figure B2.** Causal graphs detected by the causal discovery algorithm from the TOMCAT CTM simulation for the period 2004-2021 with different significance level $\alpha_{pc}$ and $\tau_{max}$=2.

Figure B1 shows the causal graphs detected by the LPCMCI algorithm for the observations (a) and the ~~lack of detection of these connections in the~~ TOMCAT CTM simulation (~~panel b) in the tropical middle stratosphere during the period 2004-2018.~~ b) for the period 2004-2021 with $\alpha_{pc} = 0.05$ and $\tau_{max} = 2$ in. For the observations, the causal discovery algorithm does not identify the direction of the $N_2O$–$O_3$ link. In addition, the $w^*$–$O_3$ connection is detected as bidirectional, indicating the presence of a latent common driver of $w^*$ and $O_3$ rather than a resolved causal direction. For the TOMCAT CTM simulation, contrary to observations, the causal discovery algorithm does not detect the anticipated negative link from $w^*$ to $N_2O$, however captures the negative contemporaneous links (without directions) between $N_2O$ and $NO_2$, and $NO_2$ and $O_3$.

Further analysis of the output from the causal discovery ~~revealed that, in observations,~~ from the observations revealed that the algorithm successfully identified a significant lagged causal link from $w^*$ to $N_2O$, indicating that past values of $w^*$ have a direct influence on $N_2O$. This relationship remained significant even after conditioning on other variables and their lags, suggesting a robust causal connection. In contrast, in the TOMCAT CTM simulation, the same lagged link was initially detected but ~~was later removed after conditional independence testing. Specifically, after conditioning on~~ became statistically insignificant once $O_3$ was included in the conditioning set, indicating that $O_3$ ~~at a lag of one month, the statistical dependence between $w^*$ (lagged by one month)~~ explains most of the shared variability between transport and $N_2O$~~(at the current time step) disappeared. This implies that,~~. To further investigate the reason for the removal of one-month lagged $w^*$ to $N_2O$ link in the TOMCAT CTM simulation ~~,~~ for the period 2004-2021, Fig. B2 shows the results of causal discovery after excluding $O_3$ ~~at a one month lag explains the relationship between~~ from the variable set. In this reduced setup, the $w^*$ ~~and $N_2O$, making the direct causal link from $w^*$~~ to $N_2O$ ~~unnecessary. In this regard, we performed additional testing on the~~ one-month lagged link reappeared and was statistically significant across tested $\alpha_{pc}$. This indicates that the previously missing transport signal is not due to a lack of dynamical coupling in the TOMCAT CTM simulation~~and found that removing O~~, but rather to conditional independence induced by the strong covariance among the chemical variables. In the TOMCAT CTM simulation, $O_3$ ~~from the analyzed system and using the full dataset from 2004–2018 (without excluding dates where observational data was missing) allowed the causal discovery algorithm to detect the lagged connection between $w^*$ and~~ has strong instantaneous coupling with both $N_2O$ ~~(see Fig. B2). In particular, as shown in Fig. B2, the lagged positive connection from~~ and $NO_2$, and therefore accounts

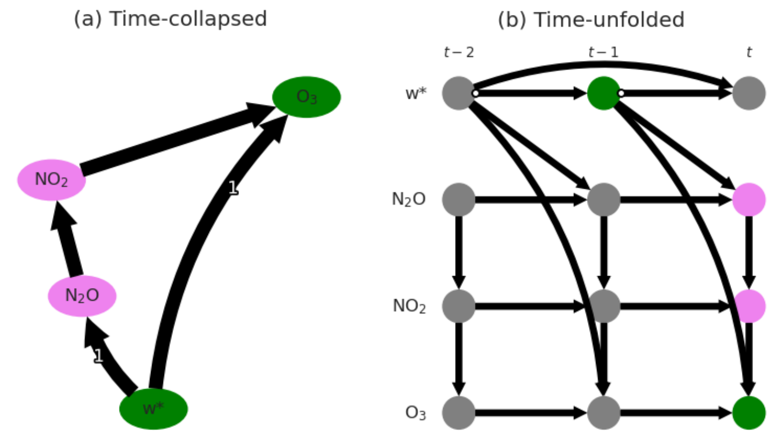

**Figure C1.** Example of the total effect of $w^*$ on $O_3$ (green) as a sum of a direct lagged link and indirect via $N_2O$ and $NO_2$ mediators (violet) in the time-collapsed graph (a)~~and time-unfolded graph (b)~~, which corresponds to the causal graph from observations shown in Fig. 4a and time-unfolded graph (b).

for a large part of their common variability. Consequently, once conditioning on $O_3$, the remaining unique contribution of $w^*$ to $N_2O$ ~~simulation appears for the significance levels ($\alpha_{pc}$) 0.2, 0.1, 0.05. This suggests that the absence of the causal link in TOMCAT may be sensitive to both the presence of~~ variability is no longer statistically detectable. The re-emergence of the $w^*$ to $N_2O$ link in the reduced system (without $O_3$~~in the model and data availability constraints.~~) therefore indicates that the transport influence is present, but in the full multivariate setup it becomes statistically hidden because the strongly coupled chemical tracers ($O_3$ and $N_2O$, and $O_3$ and $NO_2$) explain most of the same variability.

~~Causal graphs detected by the causal discovery algorithm from (a) observations (b) the TOMCAT CTM simulation for the period 2004-2018 with significance level $\alpha_{pc}$=0.02 and $\tau_{max}$=1.~~

~~Causal graphs detected by the causal discovery algorithm from the TOMCAT CTM simulation for the period 2004-2018 with different significance level $\alpha_{pc}$ and $\tau_{max}$=1.~~

## Appendix C: Total causal effects

To better understand the causal effects across different time lags, Fig. C1 depicts (a) time-collapsed and (b) time-unfolded causal graphs based on the observational causal graph detected in Fig.4~~b~~ a in the manuscript. As an example, the total effect of $w^*$ on $O_3$ (green nodes) consists of a direct path from $w^*$ to $O_3$ and an indirect path, which is mediated by $N_2O$ and $NO_2$ (violet nodes) as shown in Fig. C1a. The indirect path of the influence of $w^*$ on $O_3$ is then better represented via a time series causal graph (Fig. C1b). The total causal effect across different time lags of this relationship is shown and discussed in Fig. 7c in the manuscript.

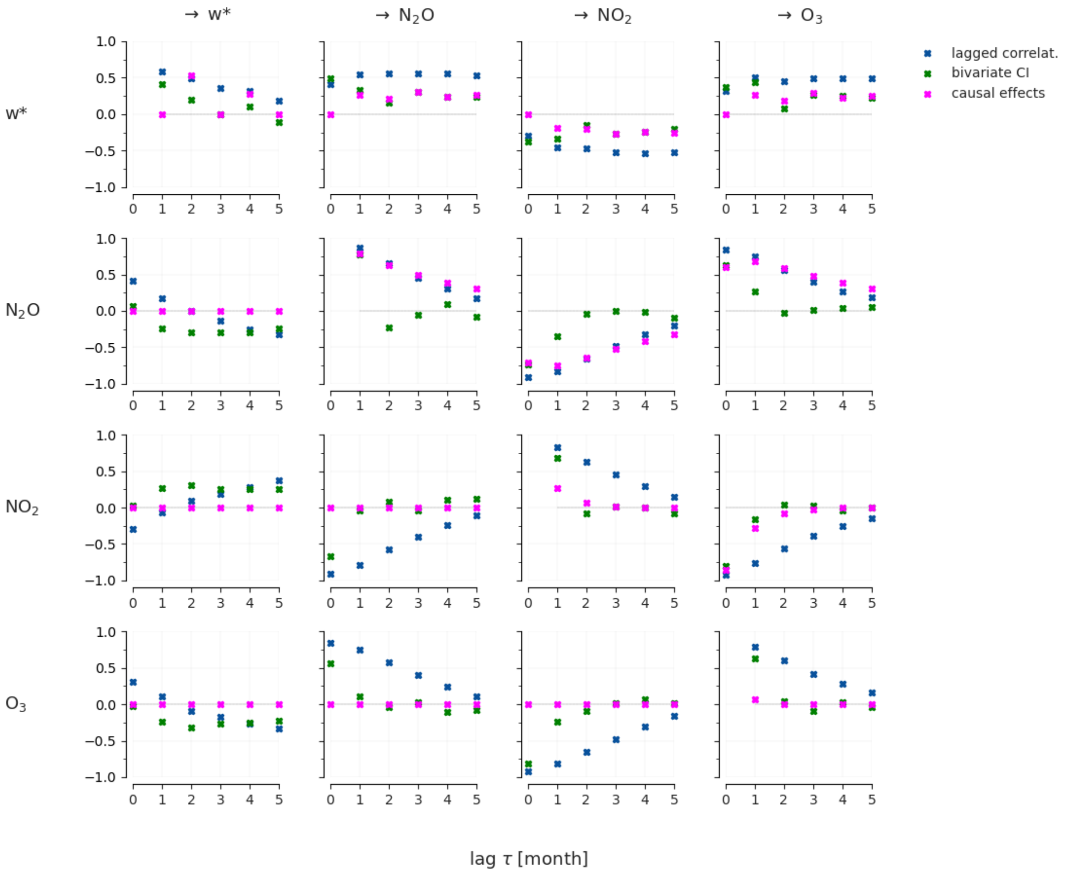

**Figure D1.** Lagged dependencies across different time lags (in months) from observations for the period ~~2004-2018~~ 2004-2021 based on partial correlations (blue), bivariate partial correlations (green), and estimated Wright coefficients (magenta).

## Appendix D: Dependencies vs. causal effects across different time lags

The lagged unconditional dependencies, such as the lagged correlations based on partial correlation, are helpful to identify the maximal time lag $\tau_{max}$ to choose in the causal discovery algorithm. However, large autocorrelation might inflate lag peaks Runge et al. (2014). To condition out some part of the autocorrelation, the bivariate, lagged CI test (partial correlation as an example) can be applied. The comparison between the lagged dependencies from both tests is shown in Fig. D1. We also plot lagged dependencies based on the calculated Wright coefficient (labeled as ’causal effects’), which directly indicates the strength of causal relationships and takes into account the direction of causal influence (see, e.g. Fig. 4 in the manuscript). For example, the causal effects in the last row in Fig. D1 are all zero since $O_3$ is not a cause of any analyzed variable in the system.

*Author contributions.* E.G. performed the analysis, prepared all figures, and led the writing of the manuscript. J.R. developed the causal discovery tool that supported this study. E.G. developed the code for data pre- and post-processing and implemented a causal inference workflow using J.R.'s package and code from Fons et al. (2023). M.P.C., S.S.D., and W.F. designed and performed the TOMCAT CTM simulation. All co-authors commented on the initial and revised drafts of the manuscripts and contributed to the interpretation of the results.

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