

Quantifying past, present and future **Stratospheric and Tropospheric Ozone over the Alps and Europe (STOA)**.

## 1. Research plan

### 1.1 Current state of research in the field

Atmospheric ozone, located mainly in the stratosphere, acts as a shield to prevent harmful solar ultraviolet radiation from reaching Earth's surface. Total column ozone (TCO) represents the total content of ozone in the atmosphere and is defined by the vertical integration of the ozone concentration in all atmospheric layers. TCO has often been used as a proxy for the amount of ozone contained in the stratosphere. However, observing changes in TCO cannot be automatically attributed to stratospheric ozone changes, since, although only about 10% of the column resides in the troposphere, absolute changes in the observed TCO could either have occurred in the troposphere or in the stratosphere, or partitioned between both.

The significant ozone decline, due to halogen containing ozone depleting substances (ODSs), observed in the last century led to the ratification of the Montreal Protocol and its amendments to phase out ODSs and thereby allow for an eventual recovery of the ozone layer to pre-1960s levels. Due to the decadal time scales involved in the observed ozone depletion and the eventual expected recovery, long-term measurements of atmospheric ozone are necessary. Ground- and satellite-based measurements of TCO, as well as ozone profiling by ozone soundings, satellite limb sounders, microwave radiometers, and LIDARs are being performed to monitor the state of the atmospheric ozone layer and quantify its evolution and its eventual recovery. To evaluate decadal changes requires that datasets from different instruments are carefully merged using overlaps over long periods, to account for individual instrument biases and uncertainties (degradation). This, in principle, should lead to a homogenised dataset that can be used to assess changes in the ozone layer over decadal time scales. As has been shown by Weber et al. (2022), small positive trends in TCO have been estimated, and could be linked to the decreasing trends in ODSs. However, climate changes also affect the recovery of ozone through the greenhouse gas (GHG) impacts on the composition, temperature, and dynamics of the stratosphere (Keeble et al., 2021; Kult-Herdin et al., 2023). While the radiative-chemical effects can be relatively easily identified and modelled, the dynamical component (e.g., changes in the Brewer-Dobson circulation) are more difficult and require chemistry-climate models as they provide a self-consistent representation of dynamical aspects of climate and their coupling to the ozone chemistry (Shepherd, 2008).

The current understanding of atmospheric ozone recovery has been summarised as follows in the most recent WMO scientific assessment on ozone depletion (WMO, 2022):

- Outside polar regions, upper stratospheric ozone trends of 1.5-2.2%/decade have been observed.
- Models project a small recovery in the lower-stratospheric ozone while observations suggest small decreases, which is more evident in the Northern Hemisphere (NH).
- Attribution of TCO trends requires knowledge of changes in ozone in both the troposphere and stratosphere.
- Current and future tropospheric ozone changes may contribute significantly to TCO changes or offset stratospheric ozone changes.

The importance of quantifying tropospheric ozone changes has been considered as essential in WMO (2022) to understand the drivers of the TCO trend and get consistency between the total column ozone and profile data. European column ozone measurements are the densest and longest on a global scale and therefore provide a unique opportunity to address issues raised by the WMO assessment report regarding the past evolution and ongoing recovery of TCO. Changes in TCO are driven by both changes in the stratospheric and tropospheric partial columns, which are not individually monitored and have different drivers and different biases in global models. The European sector is furthermore of special interest as it is located at the latitudes where the stratospheric trends are most uncertain (WMO, 2022; Ball et al., 2018; 2020) and European tropospheric

ozone trends have changed sign in the recent decade(s) (Chang et al., 2021). STOA proposes to provide a holistic assessment of ozone evolution over the European domain, and the Alpine region in particular (due to its unique characteristic (altitude, air quality) and observational records), through a unique and innovative combination of observational analyses and multi-scale ensemble modelling. STOA addresses three main science goals (SG), which are discussed in detail further below:

SG1 Quantify the total and stratospheric column ozone trends from the homogenised Arosa/Davos time series and other European datasets.

SG2 Retrieve ozone profile trends at different pressure levels from the troposphere to the upper stratosphere from the homogenised ozone profile datasets merged using data from different remote sensing and in-situ platforms.

SG3 Quantify the atmospheric dynamics and chemistry contributions to the observed ozone changes and estimate the future ozone recovery from a multi-scale modelling system comprising a global CCM and regional CTM(s).

### **SG1 Total and stratospheric column ozone trends**

Since the mid-90s, TCO has remained relatively constant, with substantial year-to-year variability though, but still about 3.5% below the 1964-1980 reference mean in the NH (Weber et al, 2021). Ball et al., (2017; 2018; and 2019) have developed and applied new statistical tools to homogenise and merge satellite-based datasets, the so-called Bayesian Integrated and Consolidated Composite (BASIC) approach. The trend analyses discussed in these studies were performed using dynamical linear modelling (DLM; Laine et al., 2014; Alsing 2019). As highlighted by Ball et al. (2018) and Gaudel et al. (2018), the near-zero trends in TCO over the last 20 years can be explained by the increasing tropospheric ozone concentration that compensate for the still ongoing decline in stratospheric ozone. On the other hand, Chipperfield et al. (2018) showed that large year to year variations in the lower stratospheric ozone are dynamically driven and mask long-time changes in the lower stratospheric column as suggested by Ball et al. (2018). Furthermore, current generation global models, while agreeing on TCO trends, show compensating biases in the tropospheric and stratospheric columns, given their different respective dominant drivers. Therefore, as proposed by WMO (2022), further research into the partial column trends and their contribution to TCO is required. Furthermore, the two regions are strongly coupled to each other not only through the dynamical activity of the atmosphere, but also through the stratosphere-troposphere exchange. Modelling studies predict that changes in the Brewer Dobson Circulation (BDC) will enhance the transport of ozone from the stratosphere to the troposphere and lead to small but significant changes in tropospheric ozone, of up to several % (Neu et al., 2014; Wang and Fu, 2023). For the Arosa/Davos time series this would represent about 50% of the expected ozone recovery from the observed stratospheric ozone depletion since the 1980's (see Figure 1 below).

### **SG2 Ozone profile trends from the troposphere to the upper stratosphere**

The LOTUS project (SPARC, 2019) re-evaluated the satellite and ground-based ozone profiles data records as well as the time series analysis methods commonly used to derive long-term trends. A combined trend profile from satellite measurements showed a significant ozone decline in the pre-1997 period for all three broad latitude bands of  $-5.9\% \pm 1.9\%$  per decade in the upper stratosphere. Post-2000 trends are significantly positive in the upper stratosphere for these latitude bands, with the largest increase seen in the NH mid-latitudes with  $\sim 2.2\%/decade$  at 40 km (Godin-Beekmann et al, 2022). In the lower stratosphere, negative ozone trends are estimated, but not statistically significant, due to their quite large uncertainties both when measured and modelled (Arosio et al. 2019; Sofieva et al. 2021; Dietmüller et al. 2021). Moreover, observed negative trends in the lower stratosphere are not reproduced by models for NH mid-latitudes (Ball et al., 2020; Dietmüller et al., 2021; Zeng et al., 2022). In the troposphere, some studies (e.g., Gaudel et al. 2018) report a decline in ozone while others suggest a post year-2000 global positive trend of 1.5 DU/decade (Ziemke et al., 2017; Gaudel et al., 2020), with the latter being

supported by modelling studies (Zhang et al., 2021; Griffiths et al., 2021). As tropospheric ozone variations are highly temporally and spatially variable, it makes it difficult to get a complete picture of its trend and of its contribution to TCO. Therefore, the tropospheric ozone contribution to TCO has to be analysed not only in a form of a column, but it requires a detailed investigation of the profile structure and related drivers (e.g., long-range tropospheric transport, precursors, etc) and uncertainties.

### **SG3 Quantification of the atmospheric dynamics and chemistry contributions to the observed ozone changes and estimation of future ozone recovery from a multi-scale modelling system comprising a global CCM and a regional CTM.**

Modelling of the ozone evolution has achieved substantial progress over the last decades, and now there is more than a dozen of global chemistry-climate models (CCMs) that can treat stratospheric processes in detail and generally agree well with observations and with each other (Dhomse et al., 2018; Keeble et al., 2021). Some observed features, like close-to-negative trends in the lower-stratospheric northern mid-latitudes, are, however, not reproduced by the models (Ball et al., 2020; Dietmüller et al., 2021). Representation of the tropospheric ozone is usually also much less satisfactory (Revell et al., 2018; Zhang and Cui, 2022), since tropospheric chemistry is often simplified in global high-top models because of the computational constraints. Therefore, reliable estimates of changes in tropospheric composition are commonly assessed with high resolution chemistry transport models (CTMs), which include a more complete representation of chemical formation, loss, and deposition mechanisms as well as transport in the boundary layer and free troposphere (Makakis et al., 2018). Among the most widely used models are WRF-Chem and CAMx and many studies have shown their ability to hind- and forecast tropospheric and surface ozone burdens (e.g., Mar et al., 2016; Li et al., 2020; Huszar et al., 2020; Karlicky et al., 2023). Although the skill of models in both domains (global chemistry-climate and regional air quality modelling) has advanced substantially in recent decades, the combination of the two modelling methodologies and respective expertise has not been applied yet to study column ozone trends and changes. Within STOA we aim on closing this gap through a series of targeted transient and sensitivity simulations comprising a joint CCM-CTM modelling framework. Such framework needs to consider the contributions of individual drivers of ozone evolution in the stratosphere and troposphere in the recent past, and thus requires a series of sensitivity (single forcing) experiments. Given all the highlighted uncertainties above, the ensemble strategy is required to obtain robust driver contributions. Once applied and validated for the recent past, this framework can then also be transferred to study the expected future changes under several socio-economic scenarios.

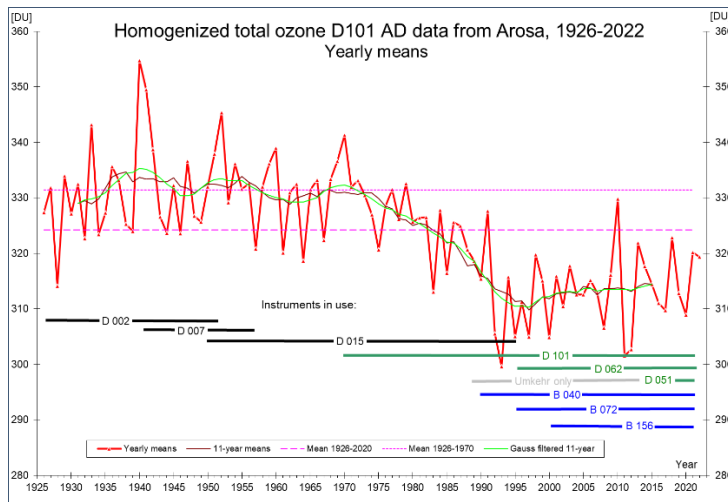
## **1.2 Current state of your own research**

### **SG1 Total and stratospheric column ozone trends**

The longest continuous TCO time series world-wide was initiated in 1926 in Arosa, Switzerland (Fig. 1), which has been one of the backbones of WMO ozone assessment reports throughout the years. Among other scientific applications, stratospheric ozone depletion over mid-latitudes was first observed using the TCO times series over Arosa due to its long measurement period extending back to pre-ozone depletion conditions, its high intrinsic homogeneity, and quality (Staehelin et al, 2018, Staehelin, 1998a, Staehelin, 1998b). As shown in our recent study by Rozanov et al. (2021), Arosa total column ozone is also highly correlated with other locations, at northern and to a lesser extent also at southern mid-latitudes, allowing results for this station to be generalised to a much wider area.

Due to the decision, made in 2010, to relocate the ozone monitoring activities from Arosa to the nearby valley of Davos, substantial efforts have been undertaken to improve current instrumentation (Stübi et al, 2021a), and develop new prototype systems to secure the TCO measurements for the next decades, to support the world-wide efforts in detecting the signs of

ozone recovery and to quantify its changes. As discussed by Staehelin et al. (2018), the Arosa ozone time series requires continuous efforts to homogenise the measurements across different instruments and periods to produce the required level of uncertainty to be used for the assessment of ozone changes and observe the eventual recovery of the ozone layer.



**Figure 1.** Homogenised total column ozone from Arosa/Davos, 1926 to 2022. 4 Dobson Instruments have been used to produce this time series over the course of the measurement period. Since the early 1990s additional Brewer spectroradiometers have been installed in addition to the Dobson instruments.

In our recent project “Investigating the future evolution of the ozone layer above Switzerland” (INFO3RS, 2018-2021), we introduced a technique for the homogenisation of TCO measurements from Brewer and Dobson spectroradiometers (Gröbner et al., 2021). It was shown that the consistency between the TCO retrieved from these instruments can be significantly improved by using measured line-spread functions and the ozone absorption cross-sections from Serdyuchenko et al. (2014). As a continuation of our previous efforts, this new methodology shall be applied to the Arosa/Davos time-series to produce a fully homogenised dataset of the highest quality that can serve as a benchmark dataset for model and satellite validation, as well as for supporting the observation and quantification of the expected ozone recovery over northern mid-latitudes. In the Alps and wider European sector several other long-term TCO monitoring sites are located. One of those is Sonnblick Observatory (Austria), operated by the BOKU Team (Fitzka et al., 2014) and widely included in international assessments and scientific studies (Eleftheratos et al., 2022; McKenzie et al., 2019). This site as well as several others contributing to the global monitoring network and reporting data to the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) will be included in the observational analyses of STOA.

## SG2 Ozone profile trends from the troposphere to the upper stratosphere

Ozone profiles have been measured at the Aerological station of the Federal Office of Meteorology (MeteoSwiss) at Payerne since the 1960's by ozone sondes launched on meteorological balloons and ground-based microwave radiometry (SOMORA MWR) since 2000. In addition to the TCO, Dobson and Brewer spectrophotometers have been measuring ozone profiles above Arosa/Davos during sunrise and sunset using the Umkehr method since 1956. This dataset is the longest ozone profile time series in the world. Together, the three observation techniques allow to retrieve ozone profiles from ground up to the lower mesosphere.

### Ozone sondes

Regular ozone soundings have been carried out in Switzerland since November 1966. Three ozone profiles a week are measured from the ground up to about 30 km, with a vertical resolution of 150 m and an accuracy of 5% in the stratosphere and 10% in the troposphere. A homogenization and a re-evaluation of the time series has been performed in order to take into account the sensor composition change (Deshler, 2017), the ozone sonde type change (Stübi, 2008), the launch time changes,

and the changes of the meteorological sonde (Jeannet, 2014). More recently, the time series has been reprocessed according to the ASOPOS 2.0 recommendations (Smit et al., 2023) with corrections for the pump flow rate and a new strategy for the residual ozone column.

The derived long-term tropospheric trends were reported to be strongly positive over the 1967–1989 period by Jeannet et al. (2007). In the same study, for the 1990–2002 period, winter trends remained positive over the whole troposphere, whereas in the other seasons, trends were generally negative near the ground and shifted to zero or positive values with increasing altitude in the troposphere. A general increase in ozone is observed in the free troposphere (especially above western North America) mainly driven by strong positive trends in winter and summer (Chang et al, 2023). As discrepancies in the tropospheric ozone trend values were reported within the European radiosonde network (Gaudel et al., 2018), a datasets harmonisation has been initiated and performed. Synergies have been created between the individual station PI (as for Payerne) and the ozone sonde working group of TOAR II (HEGIFTOM). Trends of the tropospheric column are currently under investigation using different trend estimation methods (Van Malderen R., , Thompson A.M., Smit H.G.J, Thouret V., Vigouroux C., Petropavlovskikh I., Leblanc T., Stauffer R.M., Kollonige D. E., Chang K-L., Clark, H., Poyraz D., Maillard Barras E., Sauvage B., Tarasick D., Hubert D., Homogenized ground-based and profile ozone datasets from the TOAR-II/HEGIFTOM project: Methods and station trends, in prep., 2024.).

### **Microwave radiometers**

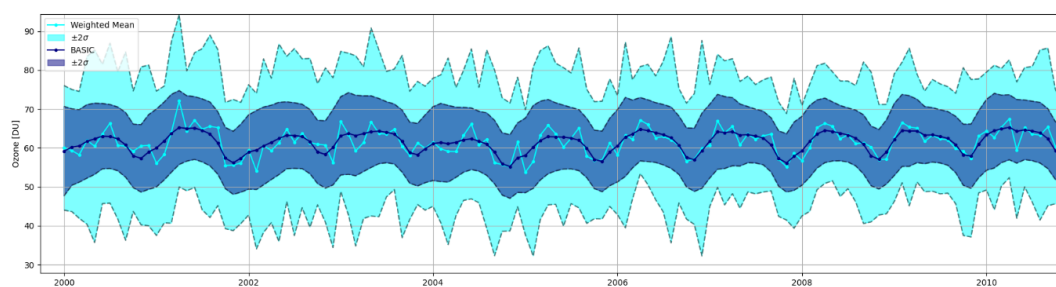
The Payerne MWR SOMORA measures the ozone volume mixing ratio in the stratosphere and lower mesosphere with a vertical resolution of 8-10 km and a time resolution of 1h independently of the weather conditions (Maillard Barras et al., 2020). The MWR ozone profiles show a good agreement of  $\pm 10\%$  with satellites such as MLS, HALOE, SCHIAMACHY and GOMOS depending on the altitude, and of  $\pm 15\%$  with the CCM SOCOL for altitudes below 50 km. Ozone profiles measured by sonde and by MWR have been previously combined in a site atmospheric best estimate (SASBE) by considering the radiosonde measurement as an a priori for the MWR retrieval below 23 km (Maillard Barras et al., 2015). The calibration processes and the retrievals of the Payerne and Bern MWRs (SOMORA and GROMOS) have been harmonized (Sauvageat et al., 2023). Their relative difference lies within 10% up to 60km. This will allow a reduction of the discrepancies in the trends derived with both instrument datasets (SPARC, 2019). Stratospheric and lower mesospheric trends have been estimated by MLR on the SOMORA MWR daytime and nighttime datasets. The trend estimations are in good accordance with trend estimated on satellite overpass datasets (LOTUS SPARC, 2019) and the measurements schedule has been excluded as cause for a significant discrepancy between the trends (Maillard Barras et al, 2020).

### **Umkehr measurements**

Ozone profiles are retrieved from zenith sky measurements by Dobson and Brewer spectrophotometers at sunrise and sunset (Götz, 1934). Two ozone profiles per day are obtained from the ground up to 50 km, with a vertical resolution of 10-15 km (Petropavloskikh et al., 2005). The Dobson Umkehr dataset was reprocessed in 2008 for corrections of technical issues (Maillard et al, 2008). The re-evaluated Dobson Umkehr time-series was used by Park et al. (2013) to derive trends using functional mixed models, and in the frame of the LOTUS project (SPARC, 2019). The trends derived from this Umkehr dataset were in agreement with trends derived by other ground-based instruments for the pre-1997 period and the post-2000 period.

More recently, the Dobson Umkehr timeseries has been homogenized by taking advantage of the collocated Brewer triad Umkehr timeseries (Maillard Barras et al., 2022). A generic straylight correction has been applied as part of the homogenization (Petropavlovskikh et al, 2022). Post-2000 trends have been estimated on the newly homogenized timeseries

by Dynamical Linear Regression (DLM) and MLR. The Umkehr datasets supports the finding that post-2000 trends are positive and statistically significant. They range between 1.5% and 2.2% per decade in the upper stratosphere of the Northern Hemisphere (NH) mid-latitudes (Godin-Beekmann et al., 2022). The 2000-2020 stratospheric ozone trends derived from the ground-based and longitudinally resolved satellite records are in close agreement, especially over the European Alpine region. DLM trend analysis of the Umkehr ozone profiles reveals a post-2004 significant positive trend in the upper stratosphere and a continuous negative trend in the middle and lower stratosphere, with significance however depending on the datasets. SG2 of this project will benefit from this study and the proposed research will allow to resolve the significant discrepancy in the lower stratospheric trend estimation. The BASIC merging method is based on the Bayesian theory as is the retrieval of the microwave radiometer water vapour (Hicks, 2020) and ozone profiles (Maillard Barras, 2020), and the LIDAR Temperature profiles (Sica and Haefele, 2015). As a proof-of-concept, a BASIC composite has been derived from the two triads Umkehr datasets for the post-2000 time range. Figure 2 shows the BASIC composite time series and its uncertainty in dark blue. A clear decrease of the uncertainty is shown when compared to the time series of the datasets weighted mean in light blue. The reduced uncertainties of the BASIC composite with respect to a standard weighted mean gives confidence for an improved significance of the estimated trends.



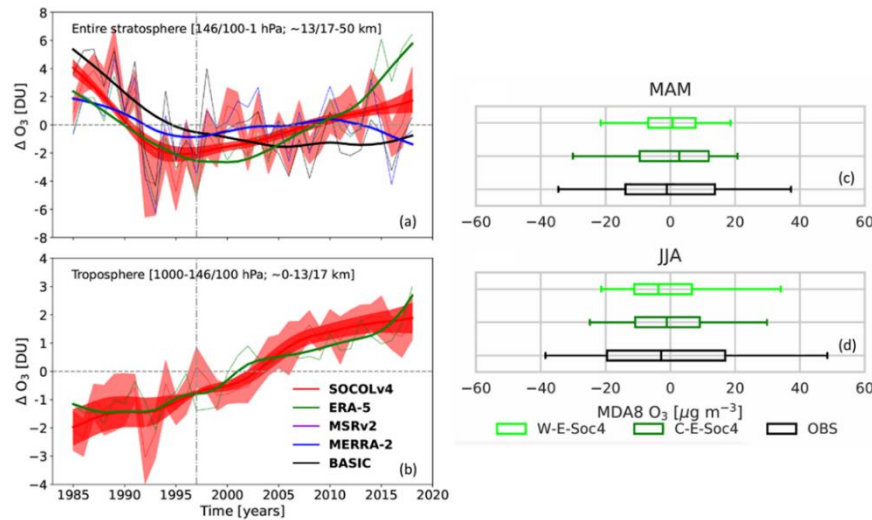
**Figure 2.** Post-2000 monthly mean time series of the BASIC composite (dark blue) and the weighted mean (light blue) of the two triads Umkehr datasets (3 Dobson and 3 Brewer at Arosa/Davos) in the middle stratosphere (62-31 hPa). The shadings show the 95% uncertainties.

### **SG3 Quantification of the atmospheric dynamics and chemistry contributions to the observed ozone changes and estimation of future ozone recovery from a multi-scale modelling system comprising a global CCM and a regional CTM.**

#### **Global Chemistry Climate Modelling using the SOCOL model at PMOD/WRC, Davos**

A global climate modelling group was established at PMOD/WRC in 2001 to support the ozone monitoring activities with ozone modelling using state-of-the-art global chemistry-climate models. For this purpose, several versions of the CCM SOCOL have been developed in collaboration with the IAC at ETH Zurich. The model has been used to simulate past, present, and future climate and ozone layer evolution driven by different natural and anthropogenic forcings. Ozone changes from 1960 until 2100 were simulated with the CCM SOCOL in the framework of the SPARC Chemistry-Climate model validation projects, CCMVal and CCMI. Results were used for the SPARC CCMVal report (SPARC, 2010), various CCMVal and CCMI papers, and the WMO assessments of ozone depletion (WMO, 2011; WMO 2018; WMO, 2022). Additionally, the evaluation of the model's performance showed good agreement with other models (Eyring et al., 2007; 2010; Dhomse et al., 2018) in simulating the historical ozone layer decline and projected recovery in the 21<sup>st</sup> century. The latest version of SOCOL is based on the Earth System model MPI-ESM and includes detailed descriptions of stratospheric and tropospheric gas-phase chemistry,

heterogeneous processes, as well as sectional model of the sulphate aerosol from previous model versions (Sukhodolov et al., 2021). The new SOCOL version has been already used to demonstrate the benefits of the Montreal ProTCool implementation (Egorova et al., 2023; Zilker et al., 2023), to analyse the consequences of a potential decline of solar activity in the future (Sedlacek et al., 2023), and to investigate the historical (Karagodin-Doyennel et al., 2022) and future (Karagodin-Doyennel et al., 2023) trends in ozone. These recent studies highlighted that the tropospheric column ozone changes can be as large as the stratospheric ones and that uncertainties are not only intrinsic to models but also to modern reanalysis products (Figure 2a-b), and that the future TCO partitioning between the tropospheric and stratospheric columns is highly scenario dependent.



**Figure 3.** Left panels (adapted from Karagodin et al., 2022): Extra-polar (55°N–55°S), annual mean evolution of partial stratospheric (a) and tropospheric (b) column ozone changes (in DU) simulated with SOCOLv4 (red) and calculated from the Bayesian BASIC ozone composite, and from the MERRA-2, ERA-5, and MSRV2 reanalyses. Thin lines represent the ozone column anomalies, while the thick lines of the same colour represent the regression model fits computed by DLM. Right panels (adapted from Karlicky et al., 2023): Normalized variability of the European maximum daily 8-hour ozone (MDA8 O<sub>3</sub>) distribution during spring and summer season in 2007-2016 for CAMx (C-E-Soc4) and WRF-Chem (W-E-Soc4) simulations driven by SOCOL output fields and observations reported to the European Environmental Agency.

### High-resolution chemistry-transport modelling at BOKU, Vienna.

The research group at BOKU focuses on chemistry-climate connections and recently several projects have been implemented with focus on tropospheric ozone changes and European ambient air quality (Huszar et al., 2020; 2021; Liu et al., 2023; Mayer et al., 2022; Karlicky et al., 2023; Staehle et al. 2022; 2023; Schmidt et al., 2023). To this end high-resolution multi-decadal time slice simulations with the CTMs WRF-Chem and CAMx have been performed. WRF-Chem is a regional, non-hydrostatic, terrain-following mesoscale model (Skamarock and Klemp, 2008) coupled with interactive chemistry (Grell et al., 2005). The model includes various gas phase chemistry and aerosol options. The gas-phase chemistry is treated using the Model for OZone And Related chemical Tracers (MOZART; Emmons et al., 2010). CAMx is an Eulerian photochemical CTM (Ramboll, 2022) and requires, as a standalone offline model, a "meteorological driver" such as WRF-Chem meteorological fields to govern transport, diffusion, deposition and chemistry. These CTMs have been successfully applied over the European and Alpine domain (e.g., Mar et al., 2016; Huszar et al., 2020; Ritter et al. 2013).



Members of the team contributed to TOAR and the recent WMO scientific assessment reports on ozone depletion (WMO, 2011; WMO 2018; WMO, 2022). Since 2021 the team of BOKU has worked together with the PMOD/WRC team on the implementation of WRF-Chem at high resolution and the use of SOCOL boundary conditions to drive CTMs. The influence of chemical and meteorological boundary conditions and CTM choice has been investigated in a recent study (Karlicky et al., 2023) showing a satisfactory representation of the ozone variability and exceedance day frequency in the European domain when CTMs are driven by SOCOL inputs (Figure 3c-d), which provides a basis for further improvement and application in STOA. The team has also made substantial progress in establishing robust bias-correction techniques for CCM or CTM output fields (Schmidt et al., 2023; Staehle et al., 2023). Furthermore, in preparation of STOA we have performed proof-of-concept modelling including the comparison of ozone profiles from the surface to 20 km with soundings at Payerne and surface ozone levels with observations from ground-based measurements. These results indicate that while the modelled ozone profiles agree quite well with the measurements in the higher part of the atmosphere, larger differences emerge closer to the surface, which will be further explored within STOA as they might be attributable to e.g., the interpolation of the rather coarse emission input data, vertical and horizontal model resolution, transboundary pollution transport.

### 1.3 Detailed research plan

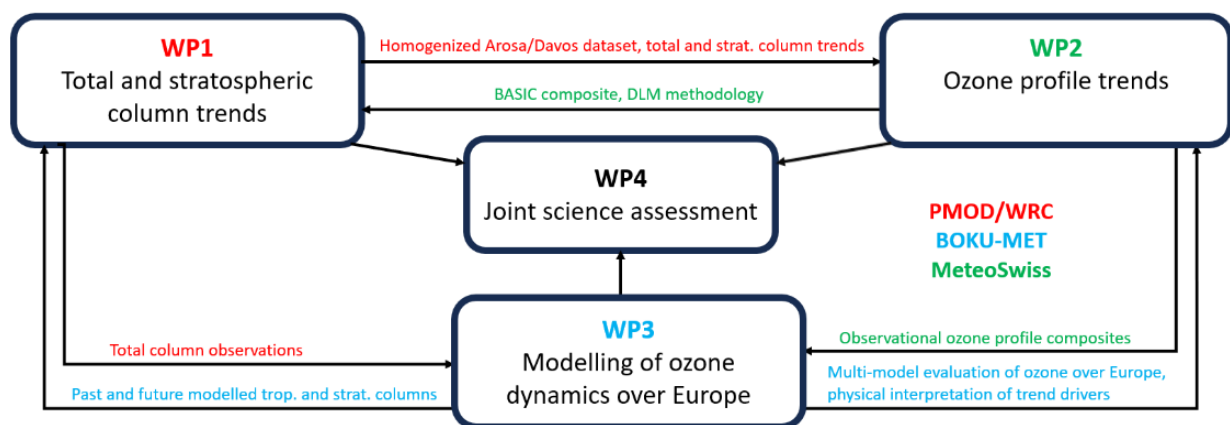
The three science goals of this project will be addressed in four work packages (see Fig. 4):

WP1 Harmonise the total column ozone measurements of Arosa/Davos from 1990 to 2022 and quantify total column ozone trends from this dataset as well as from other Alpine and European sites.

WP2 Retrieve ozone profile trends at different pressure levels from the troposphere to the upper stratosphere from the homogenised ozone profile datasets merged using data from different remote sensing and in-situ platforms.

WP3 Quantify the atmospheric dynamics and chemistry contributions to the observed ozone changes and estimate the future ozone recovery from a multi-scale modelling system comprising a global CCM and regional CTMs.

WP4 Joint science assessment by combining results from WP1, WP2, and WP3.



**Figure 4.** Schematic overview of the four work packages of STOA and the information flow between them.

The scientific analysis will be supported by applying and extending state-of-the art techniques:

- We will apply our recently developed homogenization method (Gröbner et al., 2021) to the Arosa/Davos TCO record and novel uncertainty characterization technique (Egli et al., 2022) to the set of European monitoring sites to provide a state-of-the-art observational data basis. The associated uncertainties will be derived according to the methodologies developed in conjunction with the international ozone community for Brewer and Dobson observations. PMOD/WRC has been leading



this effort through the international project EMRP ATMOZ (2014-2017), and the nationally funded project INFO3RS (2018-2021; Egli et al., 2022).

- The BASIC merging method is based on the Bayesian theory, as is the retrieval of the lidar water vapour (Hicks, 2020) and the MWR and the Umkehr ozone profiles (Maillard Barras, 2020 and 2022). The SASBE method has been used to merge ozone profiles altitude range (Maillard Barras, 2015). The BASIC composite has been successfully derived on a sample of the available dataset. The experience of Dr. Maillard Barras in the field of Bayesian methods will be used beneficially in adapting the BASIC method of Ball et al. (2017) to the ozone profile homogenisation from different measurement platforms and to train the consortium partners on this new methodology.
- The DLM approach will allow deriving long-term trends from the TCO and ozone profile datasets. The analysis will follow the Laine (Laine, 2014) and Alsing (Alsing, 2019) formalism using standard and locally specified regression components, allowing a variability of the sinusoidal seasonal modes and including the autoregressive (AR) correlation process as free parameter in the regression. The DLM methodology has been used by our consortium to estimate ozone profile trends (Maillard Barras et al., 2022).
- We will apply the global chemistry-climate model SOCOLv4 (Sukhodolov et al., 2021) and the regional chemistry-transport models WRF-Chem and CAMx to derive tropospheric and stratospheric ozone profiles over the Alpine and European domain, and validate them using available measurements at Payerne and Hohenpeissenberg and use the validated modelling results to support the analysis of the TCO dataset at Arosa/Davos, as well as other European stations. The quantification of the atmospheric dynamics and chemistry contributions and estimation of future ozone recovery for two scenarios will also be performed.

#### **WP1: Total and stratospheric column ozone trends**

The TCO measurements from the Dobson and Brewer spectrophotometers will be harmonised and reprocessed using the methodology described in Gröbner et al., 2021 (see Figure 1). This recalculation will require the use of stratospheric ozone temperature, which will be obtained from the ozone sonde dataset of Payerne and from the ECMWF re-analysis (van der A et al., 2010). Measurement uncertainties will be calculated, based on the methodologies developed in the projects INFO3RS (2018-2021), and EMRP ATMOZ (2014-2017). A homogenised TCO dataset will be derived from the data of all instruments using the BASIC methodology, using the uncertainties derived for this purpose. Total column ozone trends will then be retrieved for the period 1990 to 2020, representing the period immediately after the most pronounced ozone depletion occurred and when ozone recovery is expected to initiate. Different techniques will be applied to the harmonised datasets, such as multiple linear regression (MLR) analysis and DLM methodology which allows retrieving non-linear trends.

The derived trends will be compared with those obtained by applying the same DLM and MLR techniques to other stations in the Alps and the European domain, that extend over a sufficient time span (early 1990's to the present). Furthermore, these results will be evaluated with respect to published satellite datasets (e.g. OMI, TROPOMI, SCHIAMACHY) and published trend analyses (Weber et al., 2022). Besides observational datasets the analyses in WP1 will also comprise TCO trends from global model ensembles available from the CCMI-2022 and CMIP6 activities.

#### **WP2: Ozone profile trends from the troposphere to the upper stratosphere**

The BASIC methodology will be applied to the ozone profiles derived from ozone sondes, microwave radiometer and Umkehr measurements to merge into a representative ozone profile dataset for Switzerland. Below 30 km the composite will mostly rely on data from ozone sondes. From 30 km to 50 km, information will come from the Umkehr measurements at Arosa/Davos, performed by Dobson and Brewer spectrophotometers, as well as from the MWRs at Payerne and Bern. Despite their low

vertical resolution (10 layers from ground to 50 km), the Umkehr measurements are unique as they are available for a time period where no other measurements are available, and at altitudes which ozone sondes do not reach.

The Umkehr measurements suffer from straylight effects, which are non-negligible at high solar zenith angle. The actual retrieval does not consider these effects which induce a bias in the ozone profile (Petrovskikh, 2011). Consideration of stray light effects will be included in a revised Umkehr inversion algorithm. As total ozone measurements, Umkehr measurements are dependent of the sky conditions. Therefore, the actual need of a manual quality check, which is performed on Dobson Umkehr measurements but not on Brewer Umkehr measurements, should also be avoided and the quality insurance should be harmonized in the frame of this project. The ozone profile retrieved from Dobson and Brewer Umkehr measurements will be recalculated using the updated inversion algorithm which will include a straylight correction scheme and an automated quality check. The complete Umkehr time series of Dobson and Brewer from Arosa/Davos will be reprocessed.

Above 50 km, the composite will rely mainly on the ozone profiles from the Payerne and Bern MWRs. The merging of these datasets will take into account the different height resolutions of the retrieved ozone profiles and the uncertainty of the datasets at different altitudes ranges. While the merging of the microwave radiometers and the Umkehr ozone profiles, which show similar vertical resolutions, will simply require averaging kernel convolution (Rodgers, 2000) to degrade the vertical resolution of the microwaves ozone profiles in their common vertical range, the ozone sounding vertical resolution will be preserved by maximizing the weight of the ozone sounding measurement in the lower part of the merged profile. Furthermore, the merging process will need to handle data sets covering different time periods by adapting the uncertainties consideration scheme. In BASIC each data point will be weighted with the inverse of its uncertainty and individual offsets will be modelled for the different time periods to ensure a final time series free of steps and consistent with the individual data points and their uncertainties.

The merged ozone profile dataset will be validated against reference datasets such as the SBUV satellites datasets which are available since 1979 and show a similar vertical resolution as the Umkehr profiles (Frith, 2017), and the MLS satellite dataset which is available since 2004 (Livesey, 2008). Trends will be estimated by the DLM method on the BASIC profiles composite. The monthly means uncertainties derived by BASIC will directly weight the DLM input information in order to allow the differentiated consideration of periods in the composite time series.

Synergies with TOAR II and LOTUS III will be activated. In the framework of TOAR II, tropospheric column trends will be estimated by the quantile regression method and by MLR on ozone sonde, lidar and Umkehr datasets. The final product will provide the best fused tropospheric column trends on a global scale. LOTUS III will focus on the MLR trend estimation of ozone partial column in the stratosphere. The influence of the explanatory variables will be investigated. On one side, our project will benefit of the outputs of these studies in terms of methods and trend values comparison. On the other side, our project will bring a different merging method, the use of the DLM trend estimation method and the consideration of the specificity of the Alpine region.

### **WP3 Quantification of the atmospheric dynamics and chemistry contributions to the observed ozone changes and estimation of future ozone recovery from a multi-scale modelling system comprising a global CCM and regional CTMs**

SOCOLv4 is a state-of-the-art coupled atmosphere-ocean-aerosol-chemistry-climate model (Sukhodolov et al, 2021). It consists of the Earth System Model MPI-ESM1.2 (Mauritsen et al., 2019), the chemistry model MEZON (Egorova et al., 2003), and the sulphate aerosol microphysical model AER (Weisenstein et al., 1997, Sheng et al., 2015, Feinberg et al., 2019), with all these parts being interactively coupled. The horizontal resolution of the atmospheric part of SOCOLv4 has an approximate grid spacing of  $1.9^{\circ} \times 1.9^{\circ}$ , while the vertical resolution is set to 47 levels from the surface to 0.01 hPa. Important changes have

been recently made in SOCOLv4 and a better representation of stratospheric ozone has been achieved compared to previous SOCOL versions (Sukhodolov et al, 2021).

As a main CTM we will use WRF-Chem with a 3x3 km grid over the Alps embedded in a 9x9 km grid for the European domain. SOCOLv4 results will be used as chemical and dynamical boundary conditions for WRF-Chem. Given the specific focus on the Arosa/Davos TCO record and the complexity of ambient terrain we will reduce the grid size to 1x1 km over the area of interest (Arosa & Davos). We will run with WRF-Chem transient simulations for the historic period 1998-2019, and for selected decadal time slices over the 21st century following the representative concentration pathways and in specific setups for sensitivity studies. Furthermore, we will use a second CTM, CAMx, for sensitivity studies in case WRF-Chem ensembles do not yield statistical robustness. CAMx is a standalone offline CTM, which we will drive with WRF-Chem meteorological fields thereby assuring that differences between the two CTMs do not emerge from variability in modelled meteorological conditions. A similar combination of WRF-Chem, CAMx, and SOCOLv4 has been recently used by us in Karlicky et al. (2023). Below we introduce the four-step-simulation strategy with the joint CCM-CTM modelling framework. The list of planned simulations is presented in Table 1.

**Table 1.** Overview of SOCOL (S) and WRF-Chem (W) experiments within STOA. Note, some experiments may, dependent of robustness of the WRF-Chem ensemble, be repeated with CAMx. Acronyms for sensitivity simulations are: ODS (ozone depleting substances), GHG (greenhouse gases), sea-surface temperature (SST), sea-ice concentration (SIC), O3P (tropospheric ozone precursors).

Name of the experiment	Model with period (# of realizations)	Focus
Test	S+W2010-2015 (various)	Series of short-term runs to implement and test updates in photolysis and tropospheric chemistry modules in SOCOLv4 and to test different emission datasets in WRF-Chem
RefNudg	S1950-2019 (x1)   W1998-2019 (x1)	SOCOLv4 and WRF-Chem hindcast nudged
RefFree	S1950-2019 (x3)   W1998-2019 (x3)	SOCOLv4 and WRF-Chem hindcast free running
SenODS	S1998-2019 (x5)   W1998-2019 (x3)	SOCOLv4 and WRF-Chem with fixed ODS
SenO3P	S1998-2019 (x5)   W1998-2019 (x3)	SOCOLv4 and WRF-Chem with fixed tropospheric ozone precursors
SenGHG	S1998-2019 (x5)   W1998-2019 (x3)	SOCOLv4 and WRF-Chem with fixed GHG
SenSST	S1998-2019 (x5)   W1998-2019 (x3)	SOCOLv4 and WRF-Chem with fixed SST and SIC
FutS1	S2020-2100 (x3)   W2041-2050 (x3), W2091-2100 (x3)	SOCOLv4 and WRF-Chem future SSP1-1.9 scenario
FutS5	S2020-2100 (x3)   W2041-2050 (x3), W2091-2100 (x3)	SOCOLv4 and WRF-Chem future SSP5-8.5 scenario

**Creating boundary and initial conditions for WRF-Chem using CCM SOCOLv4:** The existing SOCOLv4 chemical output contains species from oxygen, hydrogen, nitrogen, chlorine, bromine, sulphur, and carbon groups, which are necessary for the treatment of the stratospheric ozone chemistry. The tropospheric chemistry is based on the Mainz Isoprene Mechanism (MIM), which comprises 16 organic degradation products of isoprene and 44 chemical reactions (Pöschl et al., 2000). This set of species does not contain all essential chemical species needed as an input for the RADM-2 chemical module of the applied WRF-Chem model. Thus, we will first update the SOCOLv4 chemical scheme to include the essential missing chemical species and related reactions and emissions and then rerun the model for the periods of interest. For this purpose, we will apply either updated MIM (MIM2, Taraborrelli et al, 2009) or original RADM-2 (Stockwell et al., 1990, 1997) chemical mechanisms depending on better overlap with the current WRF-Chem version. Such modification will require an update of the SOCOLv4 photolysis rate calculation module (Sukhodolov et al., 2016) to extend the list of species and to better treat cloud influence. These SOCOL modifications will

be tested in a series of short-term test runs (Test). In parallel, the pre-existing SOCOLv4 output will be used to improve the interface for data processing between the global model and the regional model. The initial simulations and model improvements will start on ETH cluster Euler then will be continued on the Swiss national supercomputing centre CSCS. Since SOCOLv4 has a coarse resolution (1.9°x1.9°) we will construct a WRF-Chem grid of high resolution (9 x 9 km) over Europe, further refined to (3 x 3 km) over the Alps with ultra-high resolution (1 x 1 km) over the Arosa and Davos region.

**Test runs with WRF-Chem:** A large part of the uncertainty in the modelling of tropospheric ozone levels arises from the uncertainty in the spatial and temporal distribution of ozone precursors (Young et al., 2018). Thus, we will perform test runs (Test), driven by different emission inventories and boundary and initial conditions stemming from the updated SOCOLv4, to compare the modelled ozone abundance with surface ozone measurements across the greater Alpine and European region (available via the reporting to the European Environment Agency) and tune the emission preprocessing using information from ground-based measurements of ozone precursors (particularly NO<sub>x</sub>) to better constrain the model. Besides surface levels a first order evaluation of the WRF-Chem tropospheric ozone profile will follow through comparison with ozone soundings available from Payerne.

**Historic multi-scale simulations and sensitivity experiments:** We will perform 3-member ensemble simulations with the updated version of SOCOL for the period of 1950-2019 (RefFree). In addition, we will also nudge the model to the ECMWF's ERA-5 reanalysis (Simmons et al., 2020) for periods from 1950 until present to get the best representativeness of chemical species and meteorology in SOCOLv4 (RefNudg). We will perform WRF-Chem simulations over Europe with a nested high-resolution domain over the Alps and an additional finer mesh over Arosa and Davos. The hind-cast simulations (RefNudg, RefFree), from the longer runs of SOCOLv4, will cover the period between 1998 and 2019, when a dense network of ground ozone observations in Europe and vertical ozone profiles from MeteoSwiss in Payerne are available. We will also evaluate WRF-Chem (and CAMx) abilities to simulate ozone by comparing it against the surface ozone observations and vertical profiles from the surface to the lower stratosphere (the BASIC ozone-profile composite previously mentioned). In addition, we will perform a series of sensitivity experiments focusing on isolating the influence of ODS (SenODS), climate change (SenGHG, SenSST), and tropospheric ozone precursors (SenO3P).

**Future multi-scale simulations:** To further investigate future ozone evolution and changes in the tropospheric column and its contribution to the total ozone column, we will run WRF-Chem for 2 decadal periods (2041-50, 2091-2100) and 2 SSPs using boundary conditions from 80-year-long transient SOCOLv4 runs (FutS1, FutS5). Simulated stratospheric and tropospheric ozone fields will allow us to investigate changes in ozone in the future compared to the past and provide estimates on the impact of future stratospheric and tropospheric ozone changes on the TCO measurements. For simulating the future, two CMIP6 emission scenario set will be used or, if already available, the new CMIP7 scenarios.

#### **WP4 Joint science assessment to achieve SG1, SG2 and SG3**

This activity will benefit from the synergy obtained by combining the expertise of the three applicants (total column ozone, ozone profiles, and ozone modelling) to retrieve past, present, and future changes in tropospheric and stratospheric ozone. The stratospheric column ozone trends will be retrieved from the harmonised total column ozone Arosa/Davos record and other European TCO time-series from WP1 by using the modelled tropospheric ozone from the multi-scale modelling performed in WP3 for the whole European area. Validation will be facilitated by the products developed in WP2. The tropospheric ozone changes for the 1998 to 2019 period will be retrieved from the multi-scale modelling results from WP3 and evaluated with respect to surface ozone measurements and other pollutants (e.g., NO<sub>x</sub>) available for this period. Finally, the results of the data analysis and the simulated future evolution of the stratospheric and tropospheric ozone will be reviewed in terms of future requirements for monitoring the stratospheric ozone layer and to derive recommendations for future monitoring activities.

## Project Consortium

### PMOD

J. Gröbner	PI, head of operational ozone measurements at PMOD/WRC. Support to ozone data homogenisation activities.
L. Egli	Ozone scientist, responsible for the quality control and quality assurance of the ozone measurements at PMOD/WRC. He will perform the uncertainty analysis for the TCO measurements of Brewer & Dobson instruments. Supported by project funds (10%).
PhDa	PhD student supported by project funds. Will be responsible for the total column ozone homogenisation of the Arosa/Davos dataset, perform total and partial column trend analyses and participate in the science assessment (WP1, WP4). Supervision by J. Gröbner and L. Harra.
T. Sukhodolov	Head of the climate group at PMOD/WRC. Expert in global chemistry-climate modelling, co-PI of the SOCOLv4 model. Will coordinate the modifications and simulations of SOCOLv4 and multi-scale model activities in collaboration with the Austrian team and supervise T. Egorova.
T. Egorova	A part-time senior scientist, supported by project funds (40%). Expert in global chemistry-climate modelling. Will focus on SOCOLv4 modifications and simulations and data preparation for regional modelling. Contribution to data analysis.
L. Harra	Professor of ETH Zurich and Director of PMOD/WRC. Formal supervisor of the two Swiss PhD students.

### MeteoSwiss

E. Maillard Barras	co-PI, scientific Staff. Responsible for ozone profiles measurements by Umkehr, microwave radiometry and by balloon-borne sonde and their data analysis. Expert in ozone data homogenisation and trend estimation. Will support the BASIC and the DLM implementation by both PhDa and PhDb.
PhDb	PhD student. Supported by project funds. Will be responsible for the ozone profile homogenisation and trend analysis (WP2). PhDb will revise the Umkehr retrieval by adding a stray light correction scheme and homogenize the Dobson and Brewer QCs. PhDb will implement the BASIC formalism for consideration of heterogeneous vertical resolutions and time scales and will take part in WP4 by interpreting the role of the tropospheric contribution in the partial column trend derived from the ozone profile measurements. Supervision by E. Maillard Barras and L. Harra.

### BOKU

H. Rieder	co-PI, Institute Head and Professor at BOKU Vienna. Expert in chemistry-climate connections with focus on ozone in the stratosphere and troposphere. Will coordinate WRF-Chem simulations, contribute to data analysis and interpretation, supervise the BOKU team, and coordinate activities with the Swiss team.
PD	PostDoc supported by project funds. Expert in chemistry-transport modelling and model output analyses and statistical data analysis. The PD will be responsible for WRF-Chem setup, testing, and the interface with SOCOLv4. PD will support the work of PhDc on CTM sensitivity simulations, model evaluation and bias correction. PD will perform CTM transient simulations and analyse CTM and CCM hindcasts projections. PD will analyze CCM sensitivity simulations and lead analyses on chemical vs. dynamical drivers of ozone changes.
PhDc	PhD student. Supported by project funds. PhD student will be responsible for performing WRF-Chem sensitivity simulations, model evaluation and output analyses and, if necessary, model bias correction. Supervision by PostDoc and H. Rieder.
I. Nadeem	Technical Staff. Expert in model setup and supercomputing architectures. Will be supporting high-performance computing on the Vienna Scientific Cluster (VSC) and post processing of model outputs on in-house BOKU HPC.

#### 1.4 Schedule and milestones

The work in WP1 will be mainly performed by PMOD.

Activity number	Date	Milestone	Staff involved
A1.1	M1 to M18	Harmonise the TCO datasets from 1990 to 2022 from Arosa/Davos from the Brewer and Dobson spectroradiometers according to the methodology described in Gröbner et al., 2021. Produce a BASIC composite for the period 1990 to 2021 with the corresponding measurement uncertainties, estimated according to the results from EMRP ATMOZ and INFO3RS.  Deliverables: 1) New harmonised reference total column ozone data sets for Brewer and Dobson instruments at Arosa/Davos (M18); 2) Peer-reviewed publication on harmonised Arosa/Davos data set (lead PhDa)	PMOD
A1.2	M18 to M24	Derive robust trend estimates of the harmonised TCO datasets of A1.1 using DLM and MLR analysis.  Deliverables: 1) Consolidated trend estimate of Arosa/Davos (M24), 2) Peer-reviewed publication on consolidated trend estimate for Arosa/Davos (lead PhDa).	PMOD
A1.3	M24 to M36	Perform TCO trend analyses from other Alpine & European stations as well as CMIP6/CCMI-2022 simulations and compare results with those for Arosa/Davos.  Deliverables: 1) Consolidated trend estimates of TOC (M36); 2) Peer-reviewed publication on consolidated European TCO trends (lead PhDa).	PMOD

The work in WP2 will be mainly performed by partner MeteoSwiss.

Activity number	Date	Milestone	Staff involved
A2.1	M1 to M18	Implementation of a stray light correction in the Umkehr retrieval and reprocessing of the complete Umkehr dataset of Dobson & Brewer. Harmonize the quality insurance processes of the raw Dobson and Brewer Umkehr datasets.  Deliverable: Peer-reviewed publication describing and validating the new retrieval with stray light correction and homogenised data set of Umkehr ozone profiles from 1956 to the present (lead PhDb).	MeteoSwiss
A2.2	M18 to M24	Construction of a BASIC composite from the ozone-sonde, the microwave radiometers, and the Umkehr Dobson and Brewer datasets with corresponding confidence intervals for the period 1956 to 2022.  Deliverables: 1) Dataset of ozone profiles from the surface to the upper stratosphere (M24); 2) Peer-reviewed publication on BASIC composite (lead PhDb).	MeteoSwiss

A2.3	M24 to M36	<p>Estimation of the stratospheric and tropospheric ozone changes by DLM and MLR from the ozone profile datasets of A2.2. Validation of the results with respect to the up-to-date literature.</p> <p>Deliverable: Peer-reviewed publication on consolidated trend estimates of ozone profiles from 1960 to present (lead PhD).</p>	MeteoSwiss
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The work in WP3 will be performed by partners PMOD and BOKU.

Activity number	Date	Milestone	Staff involved
A3.1	M1 To M12	<p>Preparation and submission of proposals to the CSCS and VSC clusters for computation hours to run SOCOLv4 and WRF-Chem/CAMx. Improvement of the interface for data processing between the global model and the regional model. Update of the chemical scheme of SOCOLv4. Test simulations with SOCOLv4 and WRF-Chem/CAMx. Hindcast simulations with SOCOLv4. Preparation of SOCOLv4 boundary conditions to run WRF-Chem and CAMx. Analysing model output fields. Prepare emission fields for historic simulations in A3.2.</p> <p>Deliverable: 1) Initial set of historic SOCOLv4 boundary conditions available for simulations and WRF-Chem/CAMx setup and input fields ready for task A3.2 (M12); 2) Peer-reviewed publication of updated SOCOL version (Lead Egorova); 3) Peer-reviewed publication on CTM sensitivity to emission inventories and other boundary conditions (lead PhD).</p>	PMOD, BOKU
A3.2	M13 to M27	<p>Historic simulations and analysis. Finalising SOCOLv4 historic runs. Performing WRF-Chem simulations using SOCOLv4 boundary conditions to calculate tropospheric ozone profiles (and columns) for the period 1998 to the present over Europe (9x9 km grid), the Alps (3x3 km grid) and over the Davos and Arosa valleys (1x1 km grid) to evaluate the potential influence of local effects on the stability of the merged total ozone record. Validation of the modelling results using the European surface ozone and TCO station network as well as the ozone sonde profiles over Payerne. Run SOCOLv4 and WRF-Chem sensitivity simulations to investigate the importance of ozone drivers over Europe to be analysed in A4.2 and A4.3.</p> <p>Deliverable: 1) Dataset of SOCOLv4 ozone fields over the historic period (M20); 2) Dataset of WRF-Chem ozone fields and evaluation of these fields between 1998-2019 (M27); 3) Peer reviewed publication describing the modelling system (lead PhD/PD); 4) All WRF-Chem and SOCOLv4 fields for A4.2 and A4.3 prepared (M27).</p>	PMOD, BOKU
A3.3	M27 to M38	<p>SOCOLv4 future simulations and analysis. Model the future evolution of the tropospheric and stratospheric ozone over the Alps and Europe using WRF-Chem in two 10-year time-slices for the period from the present to 2100 and using SOCOLv4 boundary conditions for the same period. Analysis of the results.</p>	PMOD, BOKU



		Deliverables: 1) Dataset of future SOCOLv4 ozone fields prepared (M32); 2) WRF-Chem simulation results (M38); 3) Peer reviewed publication describing the future evolution of the ozone layer under different scenarios (lead by PD).	
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The activity in WP4 will combine the work of WPs 1-3 and be a synergetic activity of all three groups. WP4 will be organised in the last two years of the project.

Activity number	Date	Milestone	Staff involved
A4.1	M36 to M48	Derive stratospheric ozone column trends for the Alpine and European region by combining the total column ozone datasets from A1.3, the profiles ozone datasets from A2.2 with the modelled tropospheric ozone profiles of A3.2 to derive stratospheric ozone column trends from 1990 to 2019 by separating the stratospheric from tropospheric ozone column changes. Validate the stratospheric ozone column trends with published trends from literature and with the trends derived in A2.3.  Deliverable: Peer reviewed publication on Alpine TCO trends (jointly lead by PhDa, PhDb and PhDc).	PMOD, BOKU, MeteoSwiss
A4.2	M24 to M48	Investigate tropospheric ozone changes in the Alpine area and European domain for the 1998-2019 period with respect to surface ozone measurements and other pollutants (e.g., NO <sub>x</sub> ) using the multi-scale modelling of WP3.  Deliverables: Two peer reviewed publications on tropospheric ozone changes and underlying drivers based on sensitivity simulations and the tropospheric contribution to changes in total column ozone content: 1) study with focus on the Greater Alpine area, 2) study with focus on the entire European domain (lead by PhDc).	BOKU, PMOD, MeteoSwiss
A4.3	M24 to M48	Estimate the contribution of atmospheric dynamics and chemistry to the observed ozone changes over the Alps and Europe using the SOCOL runs from 1990 to 2100 from A3.2 and A3.3. Specifically investigate the transport of ozone from the tropics to mid-latitudes due to the Brewer-Dobson circulation changes caused by future warming of the troposphere and cooling of the stratosphere.  Deliverable: Peer reviewed publication on dynamical and chemical contributions to ozone changes (lead PD).	PMOD, BOKU
A4.4	M36 to M48	Recommendations on requirements for future tropospheric and stratospheric ozone measurements to monitor ozone changes during the 21st century.  Deliverable: Report to monitoring infrastructure providers/funders (lead by Pls).	PMOD, MeteoSwiss, BOKU

## 1.5 Risks and Mitigation

There are no risks associated with the deliverables foreseen in WP1 and WP2. The main risk associated with WP3 concerns the significance and robustness of results and trends obtained from CCM/CTM simulations. The risk of inconclusive findings and unclear trends will be mitigated by explicitly running ensembles for sensitivity and transient experiments. Specifics on foreseen ensemble sizes are given in Table 1 of WP3. If foreseen ensembles should be too small for clean signal to noise attributions additional ensemble members will be run. Furthermore, should WRF-Chem simulations do not yield robust results/trends, simulations will be repeated with CAMx to allow robust statements via model agreement/disagreement. To avoid delays through HPC downtimes all models will be installed at both CSCS and VSC. The retrieval of stratospheric ozone trends from the total column ozone datasets using modelled tropospheric ozone columns (A4.1) will be validated at the stations with measured ozone profiles (Payerne & Hohenpeissenberg, A3.2). Activity A4.2 starts 12 months earlier than A4.1 and will compare modelling results with tropospheric ozone measurements. If the results should be not satisfactory, one mitigation activity will be to investigate the observed discrepancies and iterate the modelling activity in A3.2 to understand why these discrepancies arise and how the consistency between measurements and modelling can be improved.

## 1.6 Management

PIs Gröbner, Maillard Barras and Rieder will coordinate STOA activities at PMOD, MeteoSwiss and BOKU, respectively. The PIs will hold bi-monthly video conferences to coordinate management activities, reporting and discuss overall project progress, team performance, and achievement of milestones. The entire STOA team will meet during monthly team meetings (videoconference) and annual workshops (on site in Davos or Vienna). STOA team members will present project progress and intermediate results during team meetings and highlight potential issues and risks if they emerge. This will allow for a swift development of contingency plans and the prioritization of other STOA tasks until problems can be resolved. Individual STOA team members will meet bi-weekly with their lead PI to discuss research progress and research strategy, results obtained, drafts of scientific publications and any other scientific or administrative matters. PIs are responsible for the communication with and reporting to the funding agencies SNF and FFW. All STOA team members will report work time and tasks according to institutional and funder requirements.

## 1.7 Scientific relevance

The project provides a deep understanding of the past and expected future ozone evolution over Europe and its underlying drivers and contributions of changes in the troposphere and stratosphere to net total column changes, as well as evaluation of the uncertainties in the existing observational timeseries and measurement techniques. STOA will provide a further update and homogenization of the world's longest continuous total ozone record and thereby support and expand Switzerland's role in international assessments. We foresee that many of our results will be included in WMO, SPARC, and IPCC international assessments, specifically the next WMO assessments expected in 2026 and 2030. All STOA results will be published in open access high impact science journals, conference proceedings and presented at international conferences. The update of the SOCOL tropospheric chemistry scheme will strengthen the performance and applicability of the Swiss CCM. The set of CCM-CTM simulations will be the first of its kind and provide a comprehensive ensemble of multi-decadal transient and sensitivity experiments for the European domain and provide a plethora of further options for scientific exploitation by the STOA consortium. Last but not least, the uncertainty analyses and sensitivity tests will highlight potential deficiencies in global/regional models applied within STOA and thus indicate further areas of model improvement for the international chemistry-climate modelling community.