2005-2018 年を対象とした対流圏化学再解析データの更新

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2005-2018 年を対象とした第二世代の対流圏化学再解析を作成した。オゾン、二酸化窒素、硝酸、一酸化窒素の永積搭載センサ OMI, SCIAMACHY, GOME-2, TES, MLS, and MOPITT による観測値を同化し、1.1°の分解能で全球の再解析計算を実施した。予報モデルには MIROC-CHASER を、データ同化手法にはアンサンブルカルマンフィルターを利用し、複数化学種の濃度と排出量を同時に推定することを試みた。この手法は、対流圏の全層において様々な物質の鉛直プロファイルと季節・経年変動を再現する上で有用であった。航空機、衛星、およびオゾンゾンデなど、データ同化に利用しない独立観測データの観測との比較から、地表から下部成層圏の各層について、季節、年、十年規模の変動を現実的に捉えていることを確認した。

キーワード：データ同化、大気汚染、エミッション

1. Introduction

急速な世界経済の発展と環境政策の結果、大気汚染物質の排出は多くの地域で劇的に変化している。これらの排出量の変化は、過去数十年にわたって大気の質と気候に大きな変化をもたらしてきている。大気組成の長期的データセットは、大気環境に対する人間の活動と自然プロセスの影響を明らかにすること、大気質、人間の健康、生態系、気候に対する影響を理解することのために不可欠である。大気組成の地理・鉛直分布、時間的変動を評価するために、地上測定、オゾンゾンデ観測、衛星観測など、さまざまな観測データが利用されている。

大気組成のデータ同化では、数値モデルとして化学輸送モデルと様々な観測情報を利用することで、限られた個別の観測情報から地表濃度や放出量を含む幅広い化学成分の時空間変動を推定するのに役立つ。放出量の変動と大気組成の大気中での濃度変動を駆動するプロセスの理解のために、大気組成再解析が開発されてきている。Miyazaki et al (2015) では、アンサンブルカルマンフィルター (EnKF) データ同化技術を使用して、2005 年から 2012 年までの 8 年間を対象とした第一世代の対流圏化学再解析 (TCR-1) を構築し、さまざまな化学物質の濃度と放出を同時に推定した。 全球化学輸送モデル AGCM-CHASER (Sudo et al。、2002) および MIROC-CHASER (Watannabe et al。、2011) を予報モデルに用いた TCR-1 は、大気組成の変動性に関する包括的な情報を提供し、前駆物質の放出の変動を理解、ボトムアップ排出インベントリを評価することなどに利用されている (Miyazaki et al., 2012a, 2012b, 2014, 2017, Miyazaki and Eskes, 2017)。

本特別推進課題では、改良された EnKF データ同化システムに用いて、第二世代の対流圏化学再解析 (TCR-2) を作成した。 TCR-2 の品質は、2012 年から 2017 年の船

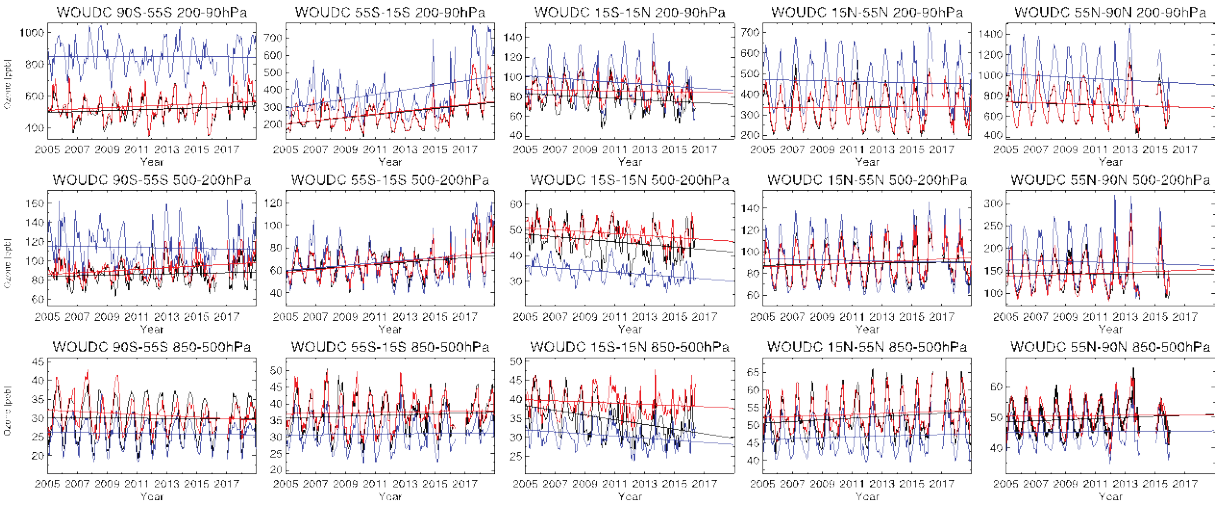

図 1 月平均オゾン濃度の時系列。オゾンゾンデによる観測（黒）、コントロールラン（青）、再解析（赤）による結果を 850-500hPa（上段）、500-200hPa（中段）、200-90hPa（下段）について示す。

舶搭載センサを用いておよび 2016 年 4 月から 5 月の KORUS-AQ 航空機キャンペーンについて、評価してきている（Miyazaki et al., 2019, Kanaya et al., 2019）。さらに、2007 年については、MOMO-Chem フレームワークを用いて様々な独立観測を用いて詳細な評価を実施している（Miyazaki et al。、2020a）。 Huijnen et al.（2019）では、オゾンのみを対象として、TCR-2 と CAMS 再解析を定量的に比較している。本研究では、表面から下部成層圏まで、毎日から十年スケールまでの、対流圏の多くの化学反応性種とエアロゾルについて、2005 年から 2018 年までの TCR-2 性能の詳細な評価結果を議論した。詳しい結果は Miyazaki et al（2020b）に記載する。

2. 手法

予測モデル MIROC-CHASER（Watanabe et al。2011）では、トレーサーの輸送、湿性および乾性沈着、および放出を計算する。対流圏と成層圏の詳細な光化学反応を含み、92 の化学種と 262 の化学反応（58 の光分解、183 の反応、21 の不均一反応）の濃度を計算する。データ同化には EnKF アプローチを採用した。複数の衛星データを用いて複数化学種の濃度と放出場を同時に最適化することにより、対流圏化学システム全体に包括的な拘束を及ぼす。この手法により、観測情報は様々な物質の間で伝搬され、多くの化学物質の寿命を修正することにも役立つ。

オゾン、NO2、CO、HNO3、および SO2 の観測値を、OMI、SCIAMACHY、GOME-2、TES、MLS、および MOPITT 衛星搭載センサから取得しデータ同化に利用した。 NOx、CO、SO2 の表面放出と稲の NOx 源、および様々な化学物質の濃度を、EnKF データ同化を使用して同時に最適化した。

3. 結果

様々な化学物質にて、独立観測データとの比較から、季節的および十年規模の変動、地表から下部成層圏まで、領域から全球規模について、再解析データの品質を検証した。オゾンゾンデ観測と比較から、再解析オゾンのバイアスは、熱帯を除く下部対流圏で 1.2 ppb 未満、南半球高緯度を除く中部および上部対流圏で 3.1 ppb 未満であった。ほとんどの地域について、相関係数は 0.85 を超た（図。1）。 TCR-1 から TCR-2 への改善は、同化に用いた観測データの改良、予測モデルのパフォーマンスと解像度の改善に起因するものと考えられる。同化により、対流圏の NO2 カラムのグローバル平均モデルバイアスは約 84～93%軽減され、人為起源およびバイオマス燃焼地域の両方において観測された季節的および年々の変動を再現した（r = 0.88-0.99）。

多成分データ同化フレームワークは、NOx、CO、SO2 の排出量の全球分布を推定することにも使用した(図 2)。化学物質の濃度のデータ同化により修正は、排出過程以外のモデル誤差から生じる観測とモデルとの不一致を軽減する上で重要な役割を果たし、排出量推定の改善に有用である。14 年間の全球排出平均値は、地表 NOx 排出量で 49.2 TgN / yr、地表 CO 排出量で 1104 TgCO / yr、地表 SO2 排出量で 35.1 TgS / yr、雷 NOx 源で 7.5 TgN / yr と推定された。これらの値は、ボトムアップインベントリに基づいて構築されたアプリオリ排出量とは大きく異なる。中国の NOx 排出量は 2005 年から 2011 年にかけて増加し、2013 年以降急速に減少し、2016 年以降増加し始めたら、中国国内での地理的な変動も大きいことが見出された。インドの NOx 排出量は、14 年間で 30%増加し続けた。米国とヨーロッパでは、環境政策により、近年 NOx 排出量の減少が見らる。 SO2 排出量は、中国（-6.1%/年）、ヨーロッパの一部（最大-6%/年）、米国東部（最大-3%/年）、日本（一方、インド（最大 5%/年）、中東（最大 4%）、メキシコ（約 4%）で大幅な増加が見られた。雷の NOx 源は、ENSO などの数年規模の気候変動に関連して、強い経年変動が見出された。これ

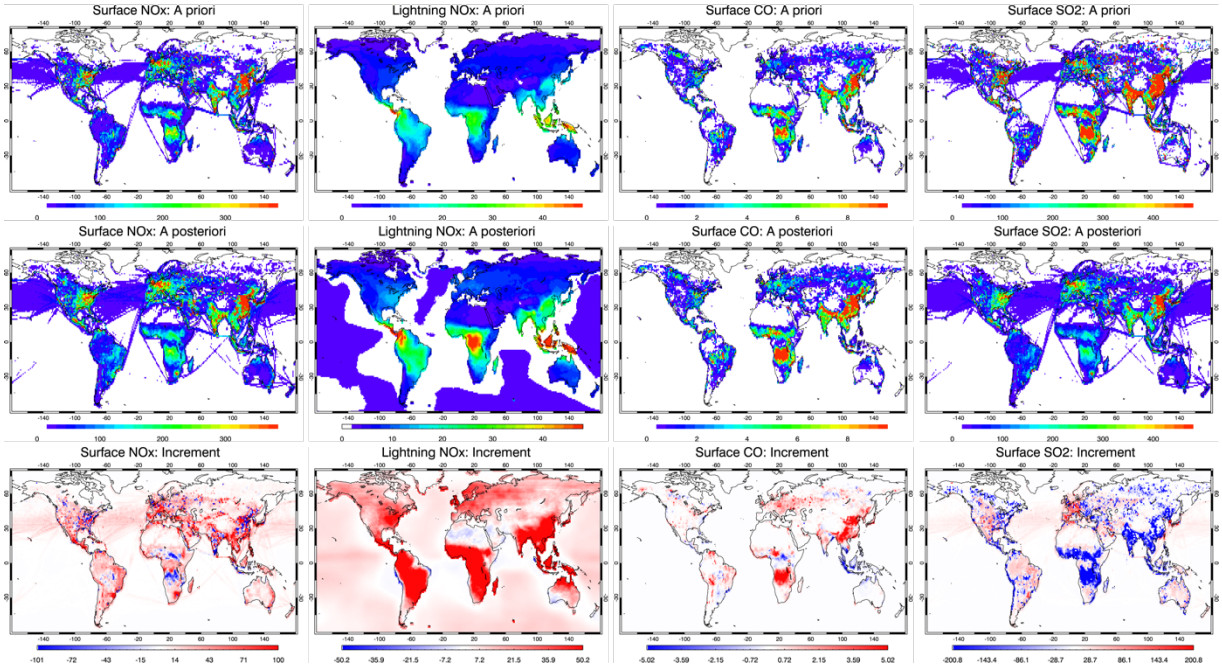

図２ 地表 NOx 排出量の全球分布（10^{-13} kgNm^{-2} s^{-1}）、（左列）、雷 NOx 源（10^{-14} kgNm^{-2} s^{-1}）（2 列目）、地上 CO 排出量（10^{-10} kgCOm^{-2} s^{-1}）（3 列目）、および地表 SO2 排出量（10^{-13} kgSm^{-2}s^{-1} で）（右列）。2005～2018 年の平均値を示す。アプリオリ値（上段）、推定値（中段）、データ同化による増分（下段）を示す。

らの結果は、大気環境のみならず、人間の健康、気候に対して、人間活動が重要な影響を及ぼしていることを示唆する。再解析データの品質には観測システムの不連続性に起因する擬似的な変動があることに注意が必要である。

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Updating Tropospheric Chemistry Reanalysis and Emission Estimates for 2005-2018

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This study presents the results from the Tropospheric Chemistry Reanalysis version 2 (TCR-2) for the period 2005-2018 at 1.1° horizontal resolution obtained from the assimilation of multiple updated satellite measurements of ozone, CO, NO2, HNO3, and SO2 from the OMI, SCIAMACHY, GOME-2, TES, MLS, and MOPITT satellite instruments. The reanalysis calculation was conducted using a global chemical transport model MIROC-CHASER and an ensemble Kalman filter technique that optimizes both chemical concentrations of various species and emissions of several precursors, which was efficient for the correction of the entire tropospheric profile of various species and its year-to-year variations. Comparisons against independent aircraft, satellite, and ozonesonde observations demonstrate the quality of the reanalysis fields for numerous key species on regional and global scales, as well as for seasonal, yearly, and decadal scales, from the surface to the lower stratosphere.

Keywords：Data assimilation, Air quality, Emissions

1. Introduction

As a consequence of rapid global economic development, along with governmental regulations, air pollutant emissions have been changing dramatically in many regions. These emission changes have led to substantial variations in air quality and climate over the past decades. A long-term record of atmospheric composition is essential to comprehend the impact of human activity and natural processes on the atmospheric environment and its effect on air quality, human health, ecosystems, and climate. Various measurements, e.g., ground-based, ozonesonde, and satellite-retrieved measurements, have been employed for assessing geographical, vertical, and temporal variations in atmospheric composition.

Chemical data assimilation can help mitigate the limitations of current observing systems using models to propagate observational information in time and space from a limited number of observed species to a wide range of chemical components, including surface concentrations and emissions. Reanalysis is a systematic approach to create a long-term data record consistent with model processes and observations, using data assimilation. To improve the understanding of emission variability and the processes controlling the atmospheric composition, chemical reanalysis products have been generated by integrating various satellite measurements. Using an ensemble Kalman filter (EnKF) data assimilation technique, Miyazaki et al (2015) simultaneously estimated concentrations and emissions of various species for an eight-year tropospheric chemistry reanalysis (TCR-1) for the years 2005-2012. The TCR-1 framework based on the AGCM-CHASER (Sudo et al., 2002) and MIROC-CHASER (Watannabe et al., 2011) models has been used to provide comprehensive information on atmospheric composition variability, understand variations in precursor emissions, and to evaluate bottom-up emission inventories (Miyazaki et al., 2012a,2012b, 2014, 2017, Miyazaki and Eskes, 2017).

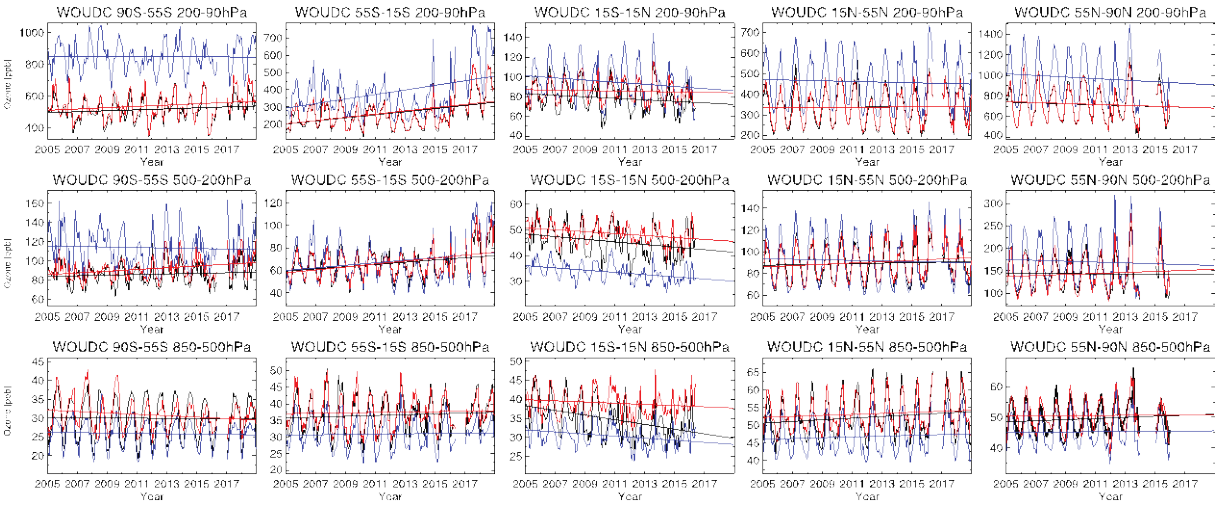

Figure 1 Time series of the monthly mean ozone concentration obtained from ozonesondes (black), control run (blue), and reanalysis (red) averaged between 850–500 hPa (upper row), 500–200 hPa (center row), and 200–90 hPa (lower row). From left to right the results are shown for the SH high latitudes (55–90°S), SH mid-latitudes (15–55°S), tropics (15°S–15°N), NH mid-latitudes (15–55°N), and NH high latitudes (55–90°N).

In this study, we produced an updated chemical reanalysis (TCR-2) developed based on an improved EnKF data assimilation system. The TCR-2 performance has been evaluated against independent observations for limited time periods in the KORUS-AQ aircraft campaign during Apr-May 2016 (Miyazaki et al., 2019) and over remote oceans using ship-borne measurements for the years 2012-2017 (Kanaya et al., 2019). The TCR-2 performance for 2007 has also been extensively evaluated against various independent observations within the MOMO-Chem framework (Miyazaki et al., 2020a). Huijnen et al. (2019) quantitatively compared the TCR-2 with operational CAMS reanalyses but for ozone only. In this study, we present the detailed evaluation results of the TCR-2 performance for the years 2005-2018 for many chemically reactive species and aerosols in the troposphere, from the surface to the lower stratosphere, at daily to decadal scales. The detailed results will be described in a future publication (Miyazaki et al., 2020b)

2. Method

The forecast model, MIROC-CHASER (Watanabe et al. 2011), contains detailed photochemistry in the troposphere and stratosphere by simulating tracer transport, wet and dry deposition, and emissions. The model calculates the concentrations of 92 chemical species and 262 chemical reactions (58 photolytic, 183 kinetic, and 21 heterogeneous reactions). Data assimilation applied here is based upon on an EnKF approach. Because of the simultaneous assimilation of multiple-species data and because of the simultaneous optimization of the concentrations and emission fields, the global distribution of various species is considerably modified in our system. This propagates the observational information between various species and modulates the chemical lifetimes of many species, as demonstrated in our previous studies. An observation operator is applied to assimilate individual measurements to map the model fields into the retrieval space.

The assimilated measurements of ozone, NO2, CO, HNO3, and SO2 were obtained from the OMI, SCIAMACHY, GOME-2, TES, MLS, and MOPITT satellite instruments. Surface emissions of NOx, CO, and SO2 and lightning NOx sources and the chemical concentrations of various species are simultaneously optimized using an EnKF data assimilation. In this framework, the improved concentrations of various species have the potential to improve the emission inversion, whereas the improved representations of emissions benefit the concentration reanalysis through a reduction in the model errors.

3. Results

The evaluation results for various species reveal the benefit of the assimilation of multiple-species data on the analysis of both observed and unobserved species profiles on both regional and global scales, for seasonal and decadal variations, and from the surface to lower stratosphere. The reanalysis ozone bias against the ozonesonde measurements was less than 1.2 ppb in the lower troposphere except for the tropics and less than 3.1 ppb in the middle and upper troposphere except for the SH high latitudes, with temporal correlations greater than 0.85 for most regions (Fig. 1). The improved agreements in TCR-2 ozone from TCR-1 can be attributed to a mixture of various upgrades, including assimilated measurements and the forecast model performance and

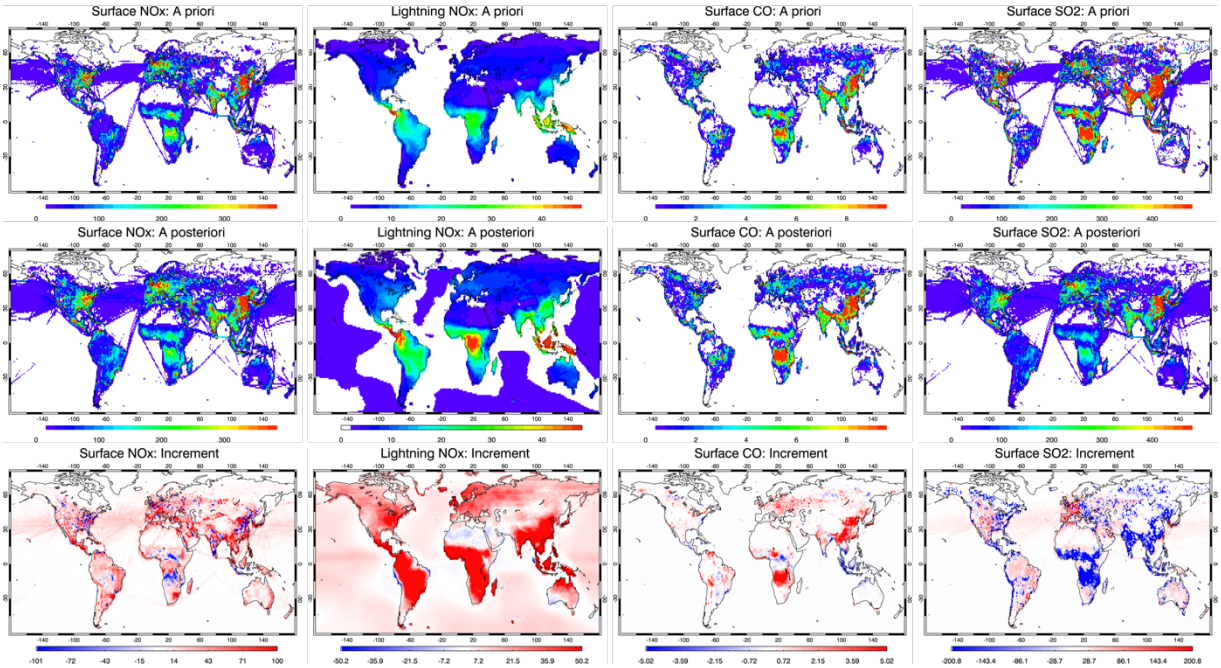

Figure 2 Global distributions of surface NOx emissions (in 10^{-13} kgNm^{-2} s^{-1}) (left columns), lighting NOx sources (in 10^{-14} kgNm^{-2} s^{-1}) (2nd columns), surface CO emissions (in 10^{-10} kgCOm^{-2} s^{-1}) (3rd columns), and surface SO2 emissions (in 10^{-13} kgSm^{-2}s^{-1}) (right columns), , and averaged over 2005–2018. The a priori emissions (upper rows), a posteriori emissions (middle rows), and analysis increment (lower rows), i.e., the difference between the a posteriori and the a priori emissions, are shown for each panel.

resolution. The assimilation also removed the global mean model biases in the tropospheric NO2 column by about 84-93%, while reproducing the observed seasonal and inter-annual changes for both industrialized and biomass burning regions (r = 0.88-0.99). The reanalysis OH shows improved agreements in global distributions over remote oceans in comparison with the ATom aircraft measurements from the surface to the upper troposphere, with the RMSE reduction of up to 30% in the free troposphere and improved north-to-south gradients. Constraints obtained for OH profiles have a large potential to influence the chemistry of the entire troposphere, which played an important role in propagating observational information among various species and in modifying the chemical lifetimes of many species. Although no aerosol observations were assimilated, improved representations of aerosols against surface in-situ measurements were obtained through corrections made to the secondary aerosol formation.

The multi-constituent data assimilation framework is also used to improve estimates of global emissions of NOx, CO, and SO2 (Fig. 2). The analysis increment produced directly via the chemical concentrations plays an important role in reducing the model-observation mismatches that arise from model errors other than those related to emissions. The global total emissions averaged over the 14 years is estimated at 49.2 TgN/yr for surface NOx emissions, 1104 TgCO/yr for surface CO emissions, 35.1 TgS/yr for surface SO2 emissions, and 7.5 TgN/yr for lightning NOx sources, which are substantially different from the a priori emissions constructed based on bottom-up inventories. Chinese NOx emissions increased from 2005 to 2011, then rapidly decreased after 2013, and then started to increase since 2016, while exhibiting substantial spatial differences within the country. Indian NOx emissions exhibit a continuous increase by 30% over 14 years. For the United States and Europe, the NOx emissions show a slowdown in NOx emission reductions in the recent years. The SO2 emissions show substantial reductions over China (by -6.1%/yr), some parts of Europe (up to -6%/yr on each grid), the eastern United States (up to -3%/yr) and Japan (up to -8%/yr), whereas strong increases are found over India (up to 5%/yr), the Middle East (up to 4%), and Mexico (about 4%), all of which are associated with environmental policies and economic activities. Lightning NOx sources exhibit strong year-to-year variability, associated with multi-year scale climate variability such as ENSO. The multi-year changes in emissions, along with the changes in meteorological conditions, led to strong increases in surface ozone over India (up to +0.25 ppb/yr) and Southeast Asia (up to +0.4 ppb/yr), as well as in tropospheric OH over the tropical western and eastern Pacific (up to +1.2%/yr) and low latitudes polluted areas (0.9-1.4%/yr) during 2005-2018. These results have strong implications on

the impacts of human activity on air quality, human health, and climate. Meanwhile, significant temporal changes in the reanalysis can partly be attributed to discontinuities in the observing systems.

4. Conclusion

The combined analysis of concentrations and emissions is considered an important development in the tropospheric chemistry reanalysis. Our comparisons suggests that improving the observational constraints, including the continued development of satellite observing systems, together with the optimization of model parameterizations, such as deposition and chemical reactions, will lead to increasingly consistent long-term reanalyses in the future. An increase in the forecast model resolution and an extension of data assimilation to aerosols are expected to improve the capability of chemical reanalysis for air quality and climate applications. Techniques to reduce the influence of discontinuities in the assimilated measurements and to employ next generation satellite retrievals would also be important developments in future chemical reanalyses. Satellite data sets from a new constellation of LEO sounders and GEO satellites (e.g., GEMS, TEMPO and Sentinel-4) will provide more detailed knowledge of ozone and its precursors for East Asia.

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