Introduction

Halogen impact tropospheric composition, notably by reducing the mixing ratios of O₃ & OH. Globally iodine chemistry has been reported to be responsible for ~10 % of gas-phase O₃ loss [Sherwen et al. 2016a] & is also a key driver of aqueous-phase O₃ loss via dry deposition to oceans [Ganzeveld et al. 2009]. However the global sea-surface iodide concentrations ([I<sub>aq</sub>]) that both these processes depend upon are highly uncertain, due to a paucity of observations. Here we present a new machine learning prediction of [I<sub>aq</sub>], incorporating new observations, and evaluate the impact on the troposphere. We also show new evidence for a change in atmospheric impacts since 1950.

Existing iodide parameterisations

Two parameterisations for [I<sub>aq</sub>] have generally been used by previous atmospheric modelling work: Chance et al. [2014] collated the 1<sup>st</sup> inventory of 925 [I<sub>aq</sub>] observations & explored correlations with 8 parameters, & found sea surface temperature (SST) squared (°C²) give the highest R² (Eqn. 1).

\[ [I_{aq}] = 0.225 \times (SST(°C))^2 + 19 \] (Eqn. 1)

MacDonald et al. [2014] used a subset of 130 (14%) observations from Chance et al. 2014 & proposed Eqn. 2.

\[ [I_{aq}] = 1.46 \times 10^6 \times \exp\left(-\frac{1960}{SST(°C)}\right) \times 1 \times 10^{-8} \] (Eqn. 2)

Fig. 1 shows the spatial prediction of these two parameterisations, and Fig. 2 compares predictions against observations. Both predict higher [I<sub>aq</sub>] with latitude, but MacDonald et al. [2014] predicts consistently ~2x higher.

New ensemble prediction

Fig. 4 shows the large reduction in the error of [I<sub>aq</sub>] prediction against observations with the new ensemble machine learning methodology [Sherwen et al. 2019].

Atmospheric impacts

We can show the effects of the new emissions with four 1-year simulations: (1) with no emissions of HOI/I<sub>aq</sub>, (2) with the new monthly [I<sub>aq</sub>] fields (3) with [I<sub>aq</sub>] fields from Chance et al. 2014 and (4) with [I<sub>aq</sub>] fields from MacDonald et al. 2014. The difference between simulation (2) and (1) gives the impacts of the inorganic emission, which depends on [I<sub>aq</sub>].

Iodine chemistry resulting from HOI-I<sub>aq</sub> emissions always leads to a decrease in ozone (Fig 6A). Compared to Chance et al. 2014, lower latitudes see higher surface ozone and high latitudes see lower ozone (Fig 6B). Compared to MacDonald et al. 2014, surface ozone is always lower (Fig 6C).

Three-fold increase in atmospheric iodine since 1950

New icecore observations [Legrand et al. 2018] show a 3-fold increase in iodine since 1950 (Fig 6). Fig. 6 also shows that this trend is qualitatively captured by the GEOS-Chem model [Legrand et al. 2018, Sherwen et al. 2017].