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Abundances of ¹⁵NO₂ and H¹⁵NO₃ in the stratosphere

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ABSTRACT

Keywords: Stratospheric chemistry Isotopic abundances Satellite remote sensing Infrared Fourier transform spectroscopy Using ACE (Atmospheric Chemistry Experiment) satellite data, the volume mixing ratios (VMRs) of stratospheric NO_2 and HNO_3 and their ^{15}N minor isotopologues have been determined, including global patterns and seasonal trends. Stratospheric $\delta^{15}N$ values for NO_2 and HNO_3 have been calculated using VMR data. $^{14}NO_2$ is relatively enhanced over the winter pole and over the equator in the lower stratosphere but at higher altitudes $^{15}NO_2$ is enhanced over the winter pole. $H^{15}NO_3$ is relatively enhanced in the lower stratosphere over the summer pole but in the upper stratosphere $H^{14}NO_3$ is enhanced.

1. Introduction

Nitrogen dioxide (NO₂) plays a key role in Earth's stratosphere; grouped together with nitrogen monoxide (NO), the two molecules are referred to collectively as NO_x [1]. The NO_x cycle is one of the primary catalytic loss cycles that destroys ozone (O₃) in the stratosphere:

$NO + O_3 \rightarrow NO_2 + O_2$	(1
	(1

 $NO_2 + O \rightarrow NO + O_2 \tag{2}$

Net reaction: $O_3 + O \rightarrow 2O_2$. (3)

Stratospheric O_3 absorbs damaging UV radiation in the 200–300 nm region and makes life possible on the surface of the Earth.

The primary source of stratospheric NO_x is N_2O , which originates from microrganisms on the surface, and is inert in the troposphere. N_2O is photolyzed in the stratosphere, and also reacts with $O(^1D)$ atoms producing NO [2,3]:

 $N_2O + h\nu \rightarrow N_2 + O(^1D) 90\%$ (4)

 $N_2O + O(^1D) \rightarrow N_2 + O_2 \, 4\%$ (5)

$$N_2O + O(^1D) \to 2NO \ 6\%$$
 (6)

NO and NO₂ are in rapid photochemical equilibrium [1]. This reaction cycle is known as the Leighton or null cycle:

$$NO + O_3 \rightarrow NO_2 + O_2 \tag{7}$$

$$NO_2 + h\nu \rightarrow NO + O$$
 (8)

$$O + O_2 + M \to O_3 + M \tag{9}$$

Net reaction: $NO + NO_2 \rightarrow NO_2 + NO.$ (10)

The NO_x cycle is terminated primarily by the formation of nitric acid (HNO₃) under daylight conditions:

$$NO_2 + OH + M \rightarrow HNO_3 + M.$$
 (11)

HNO₃ is also photolyzed back into NO₂:

$$HNO_3 + h\nu \to OH + NO_2. \tag{12}$$

 NO_2 reacts with a variety of other molecules including ClO, BrO, and HO_2 to form $ClONO_2$, $BrONO_2$ and HO_2NO_2 , respectively. At night, NO_2 reacts with O_3 to make NO_3 , which reacts with NO_2 to make N_2O_5 . All of these reactive nitrogen species are collectively called NO_y (NO, NO_2 , HNO_3 , NO_3 , N_2O_5 , $ClONO_2$, $BrONO_2$, HO_2NO_2 , etc.) molecules.

In addition to its gaseous chemistry, HNO₃ has an important role in condensed phase chemistry as a liquid and a solid (nitric acid trihydrate) in cold polar regions [4,5]. HNO₃ can form polar stratospheric clouds (PSCs), which are responsible for the destruction of ozone in the polar regions during the springtime [6].

The atmospheric nitrogen chemistry summarized above is well known and discussed in textbooks [1]. Detailed chemical transport modeling of stratospheric chemistry and dynamics generally agrees with observations. Implicit in the work discussed above, however, is the

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assumption of the "normal" isotopic composition of the reactants and products. In fact, different isotopologues such as $^{15}\rm NO_2$ and $\rm H^{15}\rm NO_3$ react (and photolyze) with different rate constants than the parent main isotopologues. These different rate constants cause isotopic fractionation, which gives additional information on atmospheric chemistry and dynamics [7]. For example, the heavy isotopologues of N₂O ($^{15}\rm NNO$, $\rm N^{15}\rm NO$ and NN¹⁸O) are relatively enriched with increasing altitude in the stratosphere, particularly over the sunlit poles, because N₂O photolysis preferentially destroys the normal light N₂O isotopologue [8].

It is customary in the area of isotopic geochemistry to use a fractional abundance δ notation [7]. For N, the primary isotope is ^{14}N with a stable minor ^{15}N isotope. In atmospheric nitrogen, the abundance ratio $[^{14}N]/[^{15}N]$ is 272 [9]; this value serves as a reference value. The $[^{14}N]/[^{15}N]$ ratio can vary for an atmospheric molecule, typically due to how the molecule formed. The magnitude of the variation from the standard $[^{14}N]/[^{15}N]$ value can be expressed in per mil by calculating the $\delta^{15}N$ value:

$$\delta^{15}N(\%) = \left[\frac{\frac{15N}{14N}_x}{\left[\frac{15N}{14N}\right]_{air N2}} - 1\right] 1000$$
(13)

with "x" denoting the sample ratio under discussion and "air N2" the standard atmospheric reference value. A positive $\delta^{15}N$ indicates the molecule has an enhancement, with the relative amount of ^{15}N larger than the standard value; $\delta^{15}N{=}0$ indicates the standard ratio is present; a negative $\delta^{15}N$ indicates the relative amount of ^{15}N present in the population is below the standard value. There is an expectation that the $\delta^{15}N$ of HNO₃ will be similar to its NO_x source [10].

Fractionation occurs in the NO/NO₂ exchange reaction, meaning a reaction in which isotopes are exchanged between molecules but no new molecules are formed [11,12]:

$$^{15}NO + {}^{14}NO_2 \leftrightarrow {}^{14}NO + {}^{15}NO_2.$$
 (14)

This type of isotopic fractionation process is known as the equilibrium isotopic effect (EIE). The EIE has been shown to be the key factor influencing the δ^{15} N value of atmospheric NO₂, and therefore HNO₃ [13–15].

There are two other isotopic fractionation effects that pertain to NO₂ [16]: the kinetic isotopic effect (KIE), associated with conventional unidirectional reactions [17] and the photochemical isotope fractionation effect (PHIFE), associated with the photolysis of NO₂. Li et al. [16] investigated fractionation of NO_x molecules in a room temperature atmospheric simulation chamber and determined that EIE enhances the $\delta^{15}N$ of NO₂ more strongly than the opposing combined effects of KIE and PHIFE, which enhance the $\delta^{15}N$ of NO. The net result of all three isotopic fractionation effects combined is the enhancement of the $\delta^{15}N$ of NO₂. They further speculated that the enhancement of $\delta^{15}N$ of NO₂ would increase at lower temperatures.

As shown by Bernath et al. [8], stratospheric N_2O has enhanced $\delta^{15}N$. Miller and Yung [18] initially proposed the enhancement was due to PHIFE preferentially depleting ¹⁴N¹⁴NO by reaction (4), which depletes N_2O in the stratosphere at rate roughly an order of magnitude greater than the other stratospheric sinks, reactions (5) and (6).

Atmospheric abundances for $\rm H^{15}NO_3$ have been reported from observations made with the MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) satellite instrument [19,20]. In the present paper, we present the experimental observations of ¹⁵N isotopic abundances for NO₂ and HNO₃ in the stratosphere from the ACE satellite mission.

2. Methods

The ACE satellite, which is also known as SCISAT, is a Canadian

mission, launched by NASA on August 12, 2003, and is still operating [21]. The orbit altitude is 650 km with the orbit inclined 73.9° to the equator. The orbital period is 97.6 min. The satellite observes the limb of the atmosphere during occultations, meaning during sunrises and sunsets, using the Sun as an illumination source for absorption spectroscopy. There are up to 30 occultations, 15 sunrises and 15 sunsets, observed in a 24-hour period. The primary instrument, which was used for this research, is a high spectral resolution (0.02 cm^{-1}) Fourier transform spectrometer (FTS), operating from 750 to 4400 cm⁻¹ ($2.2 - 13.3 \mu m$). [21]. ACE-FTS (Atmospheric Chemistry Fourier Transform Spectrometer) spectra are processed at the University of Waterloo to provide VMR (Volume Mixing Ratio) altitude profiles for 44 gaseous molecules for version 4.1/4.2 [22].

VMR data for HNO₃, H¹⁵NO₃, NO₂, ¹⁵NO₂ for v.4.1/4.2 of the ACE-FTS processing [23] were downloaded from the ACE website (https://da tabace.scisat.ca/level2/ace_v4.1_v4.2/display_data.php). The VMRs were corrected by multiplication with the HITRAN isotopologue abundances, https://hitran.org/docs/iso-meta/. The data include observations from 2004 through May 2023. The spectroscopic microwindows used in the retrievals for each of the four species are also available on the ACE website (ACE Document: ACE-SOC-0035, ACE-FTS Spectroscopy – Version 4.1). The molecular line parameters are based on values from the HITRAN 2016 molecular spectroscopic database [24], except for the ¹⁵NO₂ line parameters (not available in HITRAN 2016), which are from Perrin et al. [25].

The δ^{15} N values were calculated using Eq. (13). The VMR data and $\delta^{15} N$ values were sorted by altitude and latitude, and then placed in data bins. Each bin spans 1 km of altitude and 5° of latitude from 90° S to 90° N starting at the lower limit of the minor isotopologue retrieval (13 km at the poles and 18 km at the equator for NO2; 7 km at poles and 14 km at the equator for HNO₃) and ending at the upper limit of the retrievals (38 km for NO₂; 32 km for HNO₃). The upper altitude limits for the parent isotopologues are 59 km (polar) and 62 km (equator) for HNO3 and 52 km for NO2. The mean value of the VMRs in each data bin is taken across the entire database, separated by season (December, January February, DJF; March, April, May, MAM; June, July, August, JJA; September, October, November, SON) as indicated in the figures. Mean δ^{15} N values are calculated in the corresponding data bins from the mean H¹⁵NO₃ and ¹⁵NO₂ values. The data were filtered by removing all VMR values outside 5 standard deviations from the mean. In addition, individual ¹⁵N and ¹⁴N VMR ratios were calculated before correction by HITRAN isotopologue abundances, and all negative values and ratio values greater than 2.5 were rejected. Pixels without valid values were labelled as "NaN" and colored black in the δ^{15} N figures.

3. Results and discussion

Fig. 1 shows the VMRs of NO_2 and HNO_3 , averaged across the entire ACE database, plotted by latitude and altitude. Note stronger presence of NO_2 in the equatorial and mid-latitude region between 30 and 40 km; for HNO_3 the higher VMRs are in the polar regions near 25 km. These VMR patterns show the influence of photolysis, since HNO_3 is photolyzed to NO_2 towards the equator, but accumulates over the poles due to extended periods of darkness. The influence of photolysis can be seen more clearly by sorting the VMR data into seasonal plots (Figs. 2 and 3).

Fig. 2 shows seasonal VMR plots of NO₂. Note there is always a presence of NO₂ over the equator, as well as the summer pole. Accumulation over the summer pole begins in spring and is gone by autumn. Fig. 3 shows seasonal VMR plots of HNO₃. HNO₃ never has a significant presence over the equator. The largest HNO₃ VMRs are over the winter pole, with accumulation beginning in autumn and lingering into spring. Plots of the minor ¹⁵NO₂ and H¹⁵NO₃ isotopologues show the same patterns as the corresponding primary isotopologues.

Fig. 4 shows NO₂ δ^{15} N by latitude and altitude for the months of December, January, and February (DJF). The δ^{15} N values of up to 1000 between 15 and 20 km in Fig. 4 may be an artefact at the lower limit of



Fig. 1. Stratospheric VMR altitude-latitude distributions of NO₂ (left) and HNO₃ (right).



Fig. 2. Seasonal NO_2 VMR altitude-latitude distributions.

the retrieval. One of the main spectral interferences for $^{15}NO_2$ is H_2O and it may distort the retrieved VMR. Black pixels (NaN, not a number) correspond to NO_2 $\delta^{15}N$ without valid values. The $^{14}NO_2$ relative

enhancement over the winter pole extending to midlatitudes in the lower stratosphere, however, is real. At higher altitudes above about 30 km at the winter pole, the minor isotopologue is enhanced with $\delta^{15}N$



Fig. 3. Seasonal HNO3 VMR altitude-latitude distributions.

values up to about 600. $^{14}\mathrm{NO}_2$ is relatively enhanced in the lower stratosphere over the equator.

A similar pattern for δ^{15} N NO₂ is found over Antarctica during June, July, and August (JJA) over the winter pole, Fig. 5. The ¹⁴NO₂ relative enhancement over the equator is also present. The JJA pattern is approximately a mirror image of DJF. The spring and fall seasons show enhancements over both poles and over the equator (supplementary material).

Fig. 6 shows the HNO₃ δ^{15} N distribution for June, July, and August. The low altitude retrieval artefacts for 15 N may still be present. The minor isotopologue is enhanced in the lower stratosphere over the North (summer) pole but in the upper stratosphere the major isotopologue is enhanced. Over the equator and over the southern pole, the major isotopologue is also generally enhanced in the upper stratosphere, except over the equator. H¹⁵NO₃ shows an additional enhancement over Antarctica between 15 and 20 km that may be associated with PSCs, which occur in this altitude range during winter.

In the HNO₃ δ^{15} N plot (Fig. 7) for December, January, and February, the high altitude enhancement of the main isotopologue is present in the Antarctic (summer pole) but the minor isotopologue is enhanced in the lower stratosphere (as expected from Fig. 6). The enhancement of the minor isotopologue in the upper stratosphere over the equator is present. Over the Arctic (winter pole), the minor isotopologue is slightly

enhanced but the lower altitude PSC feature is not there. This is not unexpected since PSC occurrence in the Arctic is much less than in the Antarctic. The HNO₃ $\delta^{15} N$ plots for MAM and SON are available as supplementary data.

The ACE-FTS observations of HNO₃ isotopic abundances are generally similar to those obtained from MIPAS retrievals [20]. MIPAS has determined a global average relative abundance that is positive from 15 to 25 km (H¹⁵NO₃ is relatively enhanced from 15 to 25 km) but from 25 to 42 km the normal isotopologue is enhanced. Near the equator and at midlatitudes, the ACE distributions are negative at higher altitudes and positive at lower altitudes in agreement with MIPAS. MIPAS has also reported relative altitude profiles of H¹⁵NO₃ for equatorial, midlatitude and polar regions for the solstice and equinox time of year. For example, the MIPAS South Pole solstice (December) has a maximum δ^{15} N value of 200 at 15 km decreasing to 0 at about 22 km and reaching -700 at about 32 km and then remaining approximately constant up to 42 km. These values are in general agreement with ACE-FTS values (Fig. 7) near the South Pole for DJF.

It should be noted that ACE-FTS solar occultation measurements are always made at twilight during sunrise or sunset. For molecules like HNO_3 which have a small diurnal variation in concentration, this has a minor effect on the retrieved VMRs. However, NO_2 abundances vary more strongly between night and day so comparisons with other



Fig. 4. Altitude-latitude distribution of NO₂ δ^{15} N for Dec-Jan-Feb.



Fig. 5. Altitude-latitude distribution of NO $_2 \delta^{15}$ N for Jun-Jul-Aug.

observations and chemical models need to be made with care.

It is difficult to account for the isotopic abundance patterns observed by ACE in the stratosphere. For example, the existing experimental measurements at room temperature and atmospheric pressure for NO_2 [16] suggest that ¹⁵NO₂ should be relatively enhanced, but the observations by ACE (Figs. 4 and 5) are more complicated. The high altitude enhancement may be associated with the Brewer-Dobson circulation [26] which transports and descends stratospheric air towards the winter pole.

4. Conclusion

Near global distributions of stratospheric $^{15}NO_2$ and $H^{15}NO_3$ VMRs have been measured by the ACE-FTS and $\delta^{15}N$ distributions calculated.



Fig. 6. Altitude-latitude distribution of HNO₃ δ^{15} N for Jun-Jul-Aug.



Fig. 7. Altitude-latitude distribution of HNO $_3 \delta^{15}$ N for Dec-Jan-Feb.

 14 NO₂ is relatively enhanced over the winter pole and over the equator in the lower stratosphere. However, at higher altitudes at the winter pole, the minor NO₂ isotopologue is enhanced. The minor HNO₃ isotopologue is enhanced in the lower stratosphere over the summer pole but in the upper stratosphere the major isotopologue is enhanced. Over the equator and at midlatitudes, the main isotopologue is enhanced in the upper stratosphere. The minor HNO₃ isotopologue shows an

additional enhancement in the winter over Antarctica that may be associated with PSCs.

CRediT authorship contribution statement

P. Bernath: Conceptualization, Writing – original draft, Supervision. D. English: Formal analysis, Software, Visualization, Writing – review & editing. **C. Boone:** Data curation, Writing – review & editing. **D. Cameron:** Formal analysis, Software, Writing – original draft, Visualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data used available at https://ace.uwaterloo.ca/ after sign-up.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jqsrt.2023.108752.

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