



Retrieving Optical Depths of Nitric Acid Trihydrate Particles From IASI Measurements

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Abstract. The ozone hole is formed as a result of physicochemical processes that take place in the unique environment of the polar night, especially in the Antarctic. The polar vortex is responsible for extremely cold stratospheric temperatures, which allow polar stratospheric clouds (PSCs) to form. The present study focuses on particles of nitric acid trihydrate (NAT, $\text{HNO}_3 \cdot 3\text{H}_2\text{O}$), which is one of the key components of PSCs. We present a method to retrieve optical depths (ODs) of NAT particles from the hyperspectral measurements of the Infrared Atmospheric Sounding Interferometer (IASI) on board the Metop satellite platforms. The retrieval process is based on the construction of lookup tables using forward radiative transfer simulations for different thermal contrast situations, and is applied to 18 years of IASI spectra measured over the South Pole. The optical depth being proportional to the number of particles for a fixed particle size distribution, this results in a coherent quantitative record of Antarctic NAT particles. We show the monthly mean distributions of NAT ODs over the South Pole throughout the period 2008–2025, revealing a strong correlation with stratospheric temperatures. The distributions are more homogeneous and relevant than those derived from the previously published semi-quantitative index, as the thermal contrast is accounted for during the retrieval. In order to generalize the method, we also show its applicability over the North Pole using IASI spectra measured in January. The results reveal that IASI measurements allow the identification of events characterized by a high abundance of NAT particles on a daily basis, in both polar regions.

1 Introduction

Since the early 1980s, the ozone layer has experienced a significant annual depletion during spring over the South Pole, known as the "ozone hole" (Farman et al., 1985; Tritscher et al., 2021). Ozone molecules are converted to dioxygen molecules following catalytic cycles that involve chlorine and bromine species (Salawitch et al., 2023). One key element for the accumulation of these active halogen radicals (ClO, BrO) is the formation of polar stratospheric clouds (PSCs) (Salawitch et al., 2023; Tritscher et al., 2021). PSCs contribute both to the destabilization of the reservoir species and to the denitrification and dehydration of the stratosphere. Indeed, the conversion of chlorine and bromine species from their reservoir forms into their active radicals implies heterogeneous reactions, known to occur on the surface of PSCs. Due to their particle/droplet composition, PSCs also trap gas-phase HNO_3 and H_2O , which are therefore no longer available to neutralize the radicals (Tritscher et al., 2021). Because



of these roles, the monitoring of PSCs is essential. It is furthermore important to accurately represent the physicochemical processes at play in stratospheric models, thereby ensuring their ability to predict the evolution of the ozone hole.

In a previous work, we developed a detection method for nitric acid trihydrate (NAT, $\text{HNO}_3 \cdot 3\text{H}_2\text{O}$) particles, one important component of PSCs, from the hyperspectral measurements of the Infrared Atmospheric Sounding Interferometer (IASI) (Hermans et al., 2026b). The formation of pure NAT PSCs requires temperatures below about 195 K (denoted as T_{NAT}) in the polar lower stratosphere (4 ppmv H_2O and 10 ppmv HNO_3 at 50 hPa) (Hanson and Mauersberger, 1988). On board the Metop platforms in polar sun-synchronous orbit, the IASI sounders measure the outgoing infrared radiation of the Earth and the atmosphere between 645 and 2760 cm^{-1} , with a high spectral resolution (0.5 cm^{-1} apodized) (Clerbaux et al., 2009). Two IASI sounders are currently operational, on board Metop-B and C, launched respectively on 17 September 2012 and on 7 November 2018. The first IASI sounder, launched on 19 October 2006 on board Metop-A, has been deorbited since 2021. The next generation of hyperspectral infrared instruments is carried on the Metop-SG A-type satellites, and the first was launched on 13 August 2025. IASI-NG will ensure the continuity of the data record started by IASI with improved spectral resolution and signal-to-noise ratio (Crevoisier et al., 2014).

To go beyond detection of NAT, we present, in this work, a method to retrieve their optical depths from IASI measurements. The aim is to retrieve a quantity that is more suitable for geophysical analysis than the detection indices previously reported, given that, for a fixed particle size distribution, the optical depth is proportional to the number of particles. The retrieval method is applied uniformly to all IASI measurements over the South Pole, providing a consistent product over a period of more than 18 years (October 2007–December 2025).

The next section offers a brief recall of the detection method recently developed (Hermans et al., 2026b). The retrieval work of NAT optical depths is described in detail in Sect. 3–5. We provide with the retrieved ODs the associated uncertainties, which are estimated following the methodology described in Sect. 6. Section 7 presents results of the retrieval, with distributions of NAT optical depths over the South Pole. In Sect. 8, we address the impact of the assumptions regarding the size and shape of NAT particles on the retrieval results. We also report the extension of the method to the North Pole in Sect. 9, and, finally, we summarize the main conclusions of this work in the last section.

2 Detection method

This section only provides a review of the main elements of the detection method of Hermans et al. (2026b), which are relevant for the following. The main concept behind the detection method is the hyperspectral range index (HRI), defined, for a given IASI observation, as (Hermans et al., 2026b; Noppen et al., 2025):

$$\text{HRI} = \frac{\mathbf{K}^T \mathbf{S}^{-1} (\mathbf{y} - \bar{\mathbf{y}})}{\sqrt{\mathbf{K}^T \mathbf{S}^{-1} \mathbf{K}}} \quad (1)$$

where $\mathbf{y}$ is the measured spectrum, $\mathbf{K}$ the Jacobian (i.e., a representation of the NAT spectral signature), and $\{\mathbf{S}, \bar{\mathbf{y}}\}$ a covariance matrix and an associated mean of a set of background spectra (i.e., in absence of NAT particles). This expression, also known as the matched filter (MF), is derived from the generalized least-squares solution (Walker et al., 2011; Manolakis et al., 2016).



The HRI is a detection index quantifying the "spectral strength" of a target species over a selected spectral range. This method is particularly relevant for exploiting weak and broad features in hyperspectral measurements (Clarisse et al., 2013; Franco et al., 2018). For NAT particles, the HRIs were calculated over the 750–900 cm⁻¹ range exploiting the characteristic signature around 820 cm⁻¹ of small NAT particles (lognormal size distributions with a median radius $\lesssim 4 \mu\text{m}$). The calculation of HRIs for all measurements south of 50° S over the 17-year IASI time series (2008–2024) revealed regular detection during the austral winter (June–August), representing the first observations of NAT-containing PSCs from measurements of a passive nadir-viewing sounder (Hermans et al., 2026b).

3 Overview of the retrieval methodology

The retrieval of the optical depths of NAT particles consists in converting the HRI detection indices in optical depths (ODs). The conversion factors, called scaling factors (SFs), are hereafter determined through forward simulations of spectra and stored in lookup tables (LUTs) to enable efficient retrieval from IASI measurements. As illustrated in Fig. 1a, when considering low ODs, the HRI is directly proportional to the OD (see also Clarisse et al., 2019). A deviation from linearity becomes visible for a NAT OD above a threshold value between 0.1 and 0.2 depending on the surface temperature (Fig. 1a). As NAT particles are weak absorbers, we can assume that we remain in the linear regime. The theoretical relationship between the HRI and the OD is therefore naturally established by defining the scaling factor as the ratio of the HRI to the OD:

$$\text{SF}_{\text{LUT}} = \frac{\text{HRI}_{\text{LUT}}}{\text{OD}_{\text{LUT}}} \quad (2)$$

The HRI is, unfortunately, not only dependent on the column of NAT particles (Clarisse et al., 2019). The distributions of NAT HRIs presented in Hermans et al. (2026b) showed indeed a reduced sensitivity over the continent (particularly, over East Antarctica, see Fig. B1 in Appendix B), due to a weak thermal contrast (TC). This parameter, defined as the temperature difference between the emitting surface and the atmospheric layer of interest, is critical for the sensitivity of infrared sounders (Di Gioacchino et al., 2024). The following equation describes how the radiance emitted by a surface (at a temperature T_{surf}) is altered by its passage through an atmospheric layer, characterized by a temperature T_{atm} and an absorptivity $a(\tilde{\nu})$ (Petty, 2006; Di Gioacchino et al., 2024):

$$B(\tilde{\nu}, T_{\text{eff}}) - I(\tilde{\nu}) = a(\tilde{\nu}) [B(\tilde{\nu}, T_{\text{eff}}) - B(\tilde{\nu}, T_{\text{atm}})] = a(\tilde{\nu}) \text{TC}_r \quad (3)$$

where $B(\tilde{\nu}, T)$ is the Planck function at the wavenumber $\tilde{\nu}$ for a blackbody at the temperature T , T_{eff} is the effective brightness temperature ($B(\tilde{\nu}, T_{\text{eff}}) = \varepsilon(\tilde{\nu}) B(\tilde{\nu}, T_{\text{surf}})$, with the spectral surface emissivity $\varepsilon(\tilde{\nu})$), and $I(\tilde{\nu})$ is the outgoing radiance.

From Eq. (3), the impact of the thermal contrast (expressed in radiance, TC_r , on the right-hand side) on the intensity of the spectral signature (on the left-hand side) becomes evident. As a measure of the spectral signal strength of a target species, the HRI can be regarded as a quantitative manifestation of the left-hand side of Eq. (3), and it can therefore be deduced that, for a given absorptivity, an increase in TC will result in an increase in the HRI. This dependence is demonstrated in Fig. 1a, where an increase in the surface temperature corresponds to an increase in TC as the same atmospheric temperature profile was used for all simulations.

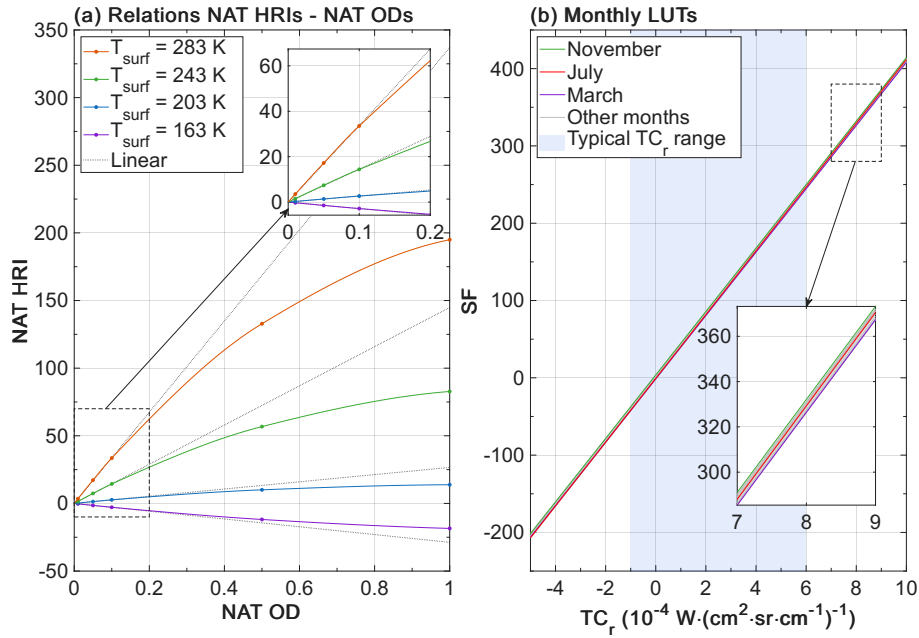


Figure 1. (a) Relations between NAT HRIs and NAT ODs (at $\lambda = 12.2 \mu\text{m}$, $\tilde{\nu} = 819.7 \text{ cm}^{-1}$) inferred from radiative transfer simulations for different surface temperatures. The colored solid lines represent interpolations between the points corresponding to forward simulations, and the dotted lines illustrate linearity. The inset provides a zoomed-in view of the low OD regime. (b) Theoretical lookup tables establishing, on a monthly basis, the relations between the scaling factors (Eq. 2) and the thermal contrast in radiance (TC_r), used to convert NAT HRIs calculated from IASI measurements over the South Pole in ODs. The LUTs for 3 selected months are shown in colors, while other months are in light gray. The inset provides a clearer view of the overlapping lines and the blue-shaded area shows the typical range of TC_r that is observed over the South Pole (latitudes higher than 50° S) throughout the year based on monthly averaged distributions.

In order to take into account the thermal contrast in the retrieval procedure, the scaling factors derived from radiative transfer simulations are stored in LUTs in relation to the corresponding TC. The HRIs calculated for measured spectra are then converted in ODs using the appropriate SF, by determining the TC in the same way as for simulated spectra.

4 Building lookup tables

The forward simulations of spectra required to build the lookup tables were executed using the 4A/OP software (Scott and Chedin, 1981). This line-by-line radiative transfer model, developed by the Laboratoire de Météorologie Dynamique (LMD), is widely used to simulate hyperspectral measurements made by infrared sounders such as IASI. It also allows to account for scattering through the implementation of different external tools (e.g., DISORT; Stamnes et al., 1988; 2000).

As explained in Sect. 3, the thermal profile is an important parameter to consider when working with infrared sounders. To best represent the profiles found in the region of the South Pole throughout the year, we chose to establish monthly LUTs



(Fig. 1b). For each month, a mean profile of the atmospheric temperatures was calculated in the region south of 50° S from the IASI L2 data (IASI-B, year 2020) and was used as input for the forward simulations by 4A/OP. To vary the thermal contrast, we simulated spectra for 7 different surface temperatures (ranging from 163 to 283 K) for each of the 12 monthly profiles. Note that we can vary the thermal contrast only through the surface temperature, because the TC is the relevant parameter for radiative transfer, rather than the two specific values of surface and atmospheric temperatures (see Eq. (3)).

For each "temperature configuration", a spectrum was simulated for a fixed NAT OD (0.01 at $\lambda = 12.2 \mu\text{m}$, $\tilde{\nu} = 819.7 \text{ cm}^{-1}$) and an equivalent reference spectrum was calculated without NAT particles. As the HRI is directly proportional to the OD for low ODs, the scaling factor is independent of the selected OD for the simulations. Consequently, simulations for different OD values are not required to build the LUTs. The reference spectrum without NAT particles replaces the mean background spectrum ($\bar{y}$) in the HRI formula for simulated spectra. This adaptation ensures that the retrieval is not impacted by systematic errors between simulated and measured spectra.

Similarly to the expression of the thermal contrast found in Eq. (3), we calculated TC in radiance (TC_r) as the difference between the Planck blackbody function (at a fixed wavenumber, $\tilde{\nu} = 800 \text{ cm}^{-1}$) at the spectrum baseline temperature (T_b) and at the atmospheric temperature of the level nearest to 50 hPa (T_{atm}), representative of the polar lower stratosphere ($\sim 20 \text{ km}$):

$$\text{TC}_r = B(800, T_b) - B(800, T_{\text{atm}}) \quad (4)$$

Here, the baseline temperature is used as a proxy for the effective temperature of the emitting surface (the land/ocean or cloud surface, given the application of the retrieval method to all-sky IASI measurements). We estimated the baseline temperature by calculating the median brightness temperature over a reduced spectral range (from 825 to 900 cm^{-1}) in order to exclude the absorption band of CO_2 . Another reason why this TC is not the true TC is that small perturbations and absorptions affect the baseline, due to, e.g., CO_2 , H_2O , HNO_3 and other trace gases. The fact that this differs from the real thermal contrast is not critical, as long as the quantity is calculated in the same way for both simulated and measured spectra.

It should be noted that NAT particles, as broadband absorbers, influence the baseline temperature of spectra. The selection of the OD value for the forward simulations could, therefore, have an impact on the LUTs because of our method for calculating the TC. We tested this hypothesis and found no significant differences between LUTs resulting from forward simulations assuming various ODs.

For the forward simulations, we took into account the following gases absorbing in the $750\text{--}900 \text{ cm}^{-1}$ range: H_2O , CO_2 , O_3 , HNO_3 , CFC-11, CFC-12, CCl_4 . The same procedure was adopted for H_2O as for the temperature profiles, i.e., we established monthly mean profiles from the IASI L2 data (IASI-B, year 2020). For CO_2 , we used a concentration of 400 ppm, which corresponds to the global average for 2015 (i.e., a year in the middle of the IASI time series). The default profiles provided by 4A/OP were applied for HNO_3 , CFC-11, CFC-12 and CCl_4 . Finally, we selected the mean O_3 profile associated with the "polar 2" class (airmass type 5) in the Thermodynamical Initial Guess Retrieval (TIGR) database (Chevallier et al., 2000), which is also supplied with 4A/OP.

The NAT particles were included at an altitude of approximately 20 km, in a layer between 60.99 and 51.53 hPa. The optical properties were previously calculated from the β -NAT (coarse mode) refractive indices of Biermann (1998) (see also Höpfner



et al., 2006) using the T-matrix method (Mishchenko and Travis, 1998). As there is no clear evidence on the size and shape of NAT particles in PSCs, due to a lack of measurements (Pitts et al., 2018), we had to decide on which parameters to use. Based on the findings of Jumelet et al. (2009), we assumed spherical particles following a lognormal size distribution, with a median radius of $1.50\ \mu\text{m}$ and a standard deviation of 1.37. We provide a more in-depth discussion on how this choice affects the retrieval in Sect. 8. It should be noted that the distribution of Jumelet et al. (2009) is not representative of the largest NAT particles, sometimes referred as "NAT rocks" (e.g., Molleker et al., 2014; Woiwode et al., 2016, 2019). We specifically chose a distribution characteristic of small NAT particles, as we know that our detection method is most sensitive to these. Indeed, the HRI relies on the distinct peak signature of the ν_2 band of NO_3^- at $820\ \text{cm}^{-1}$, which progressively decreases in intensity and disappears for larger particles ($r_g \gtrsim 4\ \mu\text{m}$) (Hermans et al., 2026b). Note also that, for the small sizes considered here, the shape of the particles is of lesser importance (see Sect. 8). In fact, the entire choice of retrieving optical depths instead of another quantity (e.g., particle number concentrations) was motivated by the uncertainty concerning the actual size and shape of NAT particles. Indeed, the OD, as a measure of the extinction, is significantly less dependent on the selected parameters (see Sect. 8).

Figure 1b presents the monthly LUTs that we obtained thanks to the forward simulations described above. We show the theoretical relations between the SFs (i.e., the ratios between the HRIs and the OD) and the thermal contrast in radiance (TC_r), for each month. The direct proportionality between these variables can be inferred from Eq. (3). It is evident from Fig. 1b that the sensitivity of IASI measurements to NAT particles is strongly dependent on the thermal contrast. If the TC is close to 0, NAT particles could be present in the optical path, but their spectral signature would not be detected with IASI. We can also observe in Fig. 1b that there are only small variations in the LUTs from one month to another.

5 Retrieval of NAT OD

As previously mentioned, the first step of the retrieval, involving the calculation of NAT HRIs for each IASI spectrum measured south of $50^\circ\ \text{S}$, has been successfully completed in a previous work (Hermans et al., 2026b). We should, however, note that the HRI data sets for IASI-B and C have now been extended to include measurements up to the year 2025. We have also improved the consistency of the HRIs throughout the years by implementing a new correction for the unexpected long-term trends observed in the HRI time series, which have been found to depend on the temperature (see Appendix A).

Selecting the appropriate scaling factor from the LUTs first requires determining the thermal contrast associated with each observation. As for simulated spectra, the baseline temperature is used as a proxy for the temperature of the emitting surface. The atmospheric temperature is extracted from the IASI L2 temperature profiles (L2 CDR for IASI-A and IASI-B up to 2022; EUMETSAT, 2022; and L2 NRT for IASI-B from 2023 onwards and IASI-C; EUMETSAT, 2017) and should, in principle, be representative of the layer in which the NAT particles are detected. As we do not have enough information to determine the exact altitude, we selected a level representative of the lower stratosphere, which is the level nearest to 50 hPa, corresponding to an altitude of $\sim 20\ \text{km}$. The choice of the level is not critical, as long as we use the same level for both simulated and measured spectra. Once the TC has been established, the scaling factor is derived from the LUTs and the optical depth at $\lambda = 12.2\ \mu\text{m}$



165 $(\tilde{\nu} = 819.7 \text{ cm}^{-1})$ is retrieved from:

$$\text{OD} = \frac{\text{HRI}_{\text{retr}}}{\text{SF}_{\text{LUT}}} \quad (5)$$

Following the retrieval, we applied a correction to account for the differences in optical path lengths as a function of the satellite zenith angle (SZA). Indeed, IASI measures spectra over a swath width of ~ 2200 km, whereas the LUTs were built for a nadir geometry. To compensate for the differences, a cosine factor was introduced:

$$170 \quad \text{OD} = \text{OD} \cdot \cos(\text{SZA}) \quad (6)$$

A post-filter was also implemented to remove clearly erroneous ODs. Indeed, in areas characterized by a high level of noise, the retrieval shows extreme values (both positive and negative), associated with low thermal contrast. The reason for this is that the scaling factors reach values close to 0 for those regions with low sensitivity (see Fig. 1b), leading to high absolute OD values (see Eq. (2)). To exclude these outliers, we developed a methodology for estimating the uncertainty associated with
 175 each individual measurement, described in the next section (Sect. 6). The retrievals associated with the highest uncertainties ($\sigma_{\text{OD}} > 0.01$) were then excluded.

Figure 2 illustrates the retrieval process for one day of IASI-B measurements. As expected, the distribution of the scaling factors (Fig. 2c) reflects that of the thermal contrast (Fig. 2b). The retrieved ODs reveal a more homogeneous distribution of NAT particles (Fig. 2d) than the initial HRIs (Fig. 2a), as the TC is accounted for during the retrieval. This is particularly
 180 evident over West Antarctica: a clear transition between the values observed over sea and over land is visible with the HRIs (high values over sea, low values over the continent), whereas the ODs are more continuous.

As visible on Fig. 2d, the post-processing essentially excludes observations over East Antarctica and over cloudy scenes, where the surface temperature is only slightly higher than the atmospheric temperature. Because of the weak thermal contrast, the sensitivity is reduced, and those observations are logically associated with the highest uncertainties (Fig. 2e). In this
 185 example, 81,573 out of the initial 278,013 observations were excluded.

It should be noted that negative OD retrievals are not excluded (Fig. 2d), despite the fact that negative values are not physically meaningful. For spectra measured in absence of the target species, the HRIs are normally distributed around a mean value of 0 with a standard deviation of 1. As a consequence, negative OD retrievals are expected for individual observations and are required to have an unbiased background distribution. Removing negative values on a single observation basis would
 190 indeed introduce a false positive bias (Clarisse et al., 2019).

The retrieval method has been applied to all IASI spectra measured south of 50° S from the 3 Metop platforms spanning a period of more than 18 years (IASI-A: October 2007–December 2020; IASI-B: March 2013–December 2025; IASI-C: September 2019–December 2025).

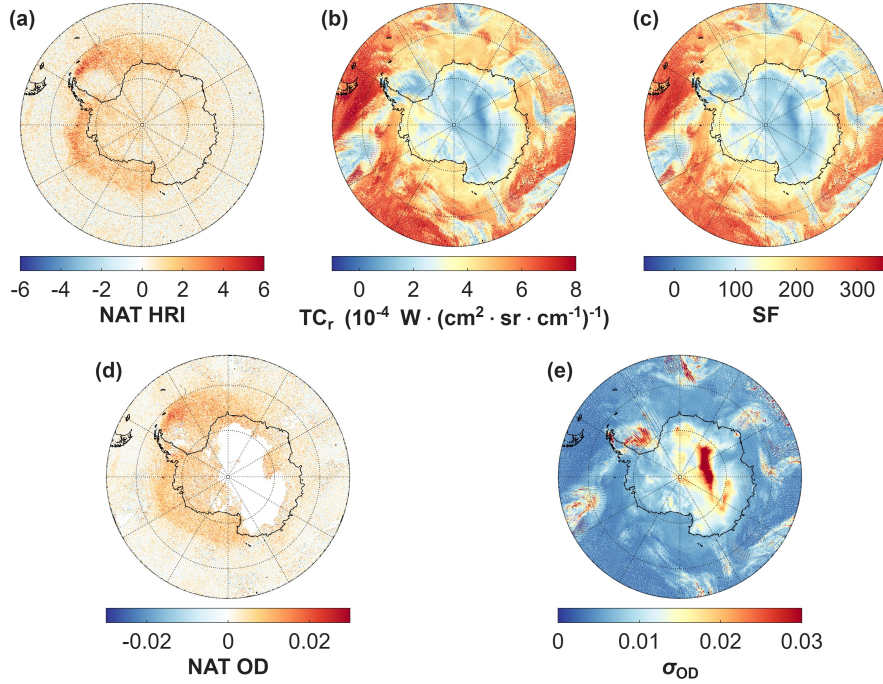


Figure 2. Illustration of the retrieval of NAT ODs from IASI-B spectra measured on 20 July 2020, with the distributions of: (a) the NAT HRIs, (b) the thermal contrast (expressed in radiance, TC_r), (c) the scaling factors (SFs), (d) the retrieved NAT ODs, and (e) the associated uncertainties (σ_{OD}). The measurements absent from the NAT OD map are excluded by the post-filter.

6 Estimation of the uncertainty

195 From the equation used to retrieve the NAT OD values ($OD = HRI/SF$), uncertainties on the ODs can be estimated by propagating the uncertainties on the two input parameters (i.e., the HRI and the SF):

$$\sigma_{OD}^2 = \left(\frac{\partial OD}{\partial HRI} \right)^2 \sigma_{HRI}^2 + \left(\frac{\partial OD}{\partial SF} \right)^2 \sigma_{SF}^2 \quad (7)$$

As SF is only dependent on the TC, this equation reduces to:

$$\sigma_{OD}^2 = \left(\frac{1}{SF} \right)^2 \sigma_{HRI}^2 + \left(\frac{\partial OD}{\partial TC} \right)^2 \sigma_{TC}^2 \quad (8)$$

200 The uncertainty on the HRI, σ_{HRI} , is 1 by definition. The partial derivative $\partial OD / \partial TC$ is estimated by introducing a small perturbation to the baseline temperature for each individual measurement. The uncertainty on the TC, σ_{TC} , is addressed by taking various contributing factors into account:

- the uncertainty on the baseline temperature due to the radiometric noise of the IASI sounder (~ 0.2 K);
- the uncertainty on the atmospheric temperature taken from IASI L2 (~ 1 K);



205 – the bias related to the use of a fixed level for the atmospheric temperature.

Given the observed range of PSC altitudes (~ 15 – 25 km) (Tritscher et al., 2021), the contribution of the latter is the dominant source of uncertainty on the TC. To estimate its value on a single observation, we considered 2 pressure levels (~ 70 and 30 hPa) in addition to the level used for the retrieval (~ 50 hPa) and assumed σ_{TC} to be the largest difference between the middle level and the upper/lower level. The observed differences range from 1 – 2 K to over than 10 K, greatly varying with the season because of the variations in the altitude of the tropopause.

Note that the data have not been prefiltered for the presence of clouds. Their influence on the TC has been taken into account by using the baseline temperature rather than the "real" surface temperature. By doing so, the thermal contrast is always the temperature difference between the emitting body (i.e., the Earth surface for a clear sky scene and the cloud surface for an opaque cloud) and the atmospheric temperature.

215 The example presented in Fig. 2 shows that the highest uncertainties are found over East Antarctica and over some cloudy scenes, where the thermal contrast reaches its lowest values. Implementing a post-filter is essential for effectively excluding the ODs affected by those large uncertainties. The remaining OD values have an average uncertainty of $6 \cdot 10^{-3}$.

7 South Pole distributions of NAT OD

To illustrate the obtained data set of NAT optical depths, we show in Fig. 3 the monthly mean distributions, on a regular grid of 1° longitude by 1° latitude, retrieved from 18 years of IASI measurements over the South Pole. Firstly, monthly distributions were produced for each year between 2008 and 2025, by averaging all available measurements (regardless of the Metop platform) in each grid cell. Then, the climatology for the period 2008–2025 was calculated based on these annual monthly distributions.

Note that if the monthly averaging resulted in negative values, an OD of 0 was assigned to the corresponding grid cells. As explained in Sect. 5, retrieval of negative ODs for individual measurements are required for an unbiased product. Here, given the large amount of data being averaged, the individual negative values should offset the positive ones, resulting in a distribution centered on 0 for grid cells devoid of NAT particles. Negative mean values reveal cases with a distribution slightly biased towards the negatives, whereas the individual ODs should average to an OD equal or greater than 0. Setting negative grid cells to 0 helps eliminate biases in the climatology by providing a more reliable estimation of the monthly distributions (Clarisse et al., 2019). Negative values were found during the months of September and October, a period for which we obtained unexpected negative HRIs, as discussed in Hermans et al. (2026b).

As expected, the NAT OD increases during the winter months (June–July–August), reaching maximum mean values around $7 \cdot 10^{-3}$ in July (Fig. 3). High ODs are found in regions surrounding the South Pole, in agreement with those where temperatures in the lower stratosphere are below the T_{NAT} value of 195 K, delimited by the blue contours in Fig. 3 for the winter months. The background values outside those cold regions are not strictly at 0 but around $1 \cdot 10^{-3}$, as already observed in May. Values over East Antarctica are noisier (or even absent) because more individual retrievals are excluded during post-processing due to lower thermal contrasts in this region.

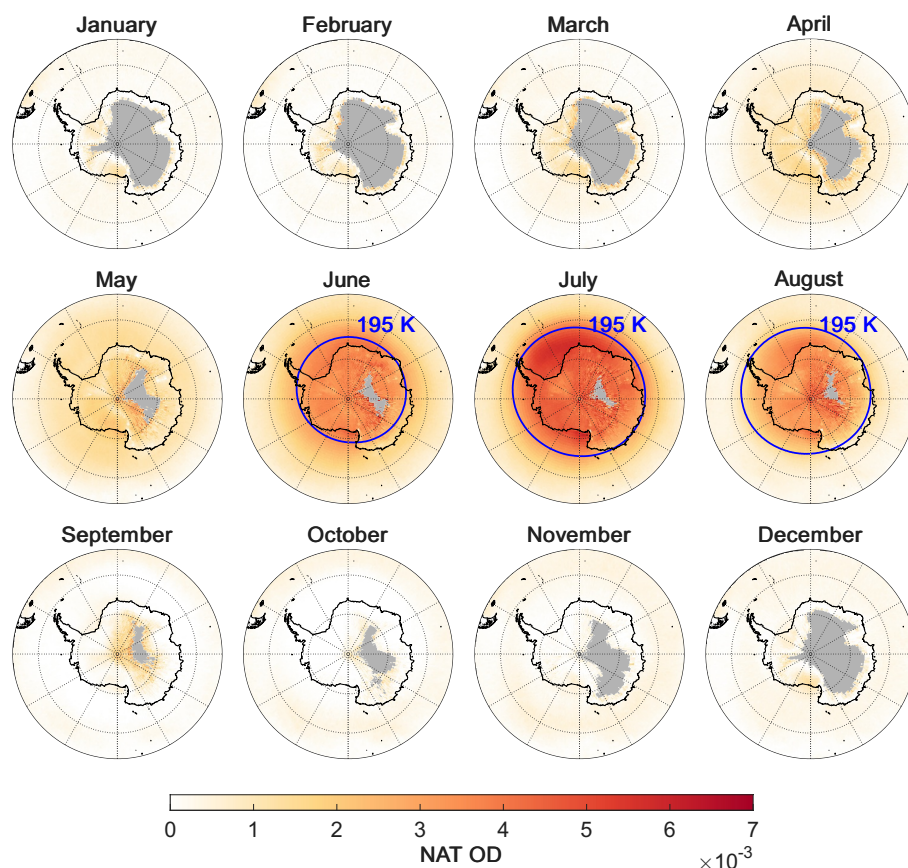


Figure 3. 2008–2025 monthly mean distributions of NAT ODs on a 1° longitude by 1° latitude grid, derived from measurements of IASI-A, B, and C. The data are first gridded monthly by year, and then an average is calculated based on these annual monthly distributions. Grid cells for which the averaging is based on less than 10 years due to the post-processing are considered as not sufficiently representative of the climatology and are shaded in gray. The blue contours for the winter months (June–July–August) mark the T_{NAT} value of 195 K at a pressure level of ~ 50 hPa.

The equivalent of Fig. 3 but for the initial detection indices, the NAT HRIs, is shown in Appendix B. The retrieval of ODs clearly shows more homogeneous distributions of NAT particles inside the 195 K contours with especially the land-sea discontinuity removed. This result demonstrates the advantage of the retrieval approach, which successfully addresses the disparity in thermal contrast between land and sea.

8 Influence of the assumed particle size distribution and shape

In this section, we discuss the choice of the particle size distribution (PSD) used to calculate the optical properties of the NAT particles. The NAT ODs described above have been obtained assuming a lognormal distribution, characterized by a median



radius of $1.50\ \mu\text{m}$ and a standard deviation of 1.37 (Jumelet et al., 2009). However, a test was conducted to determine the impact of this assumption on the retrieved OD values from IASI-B spectra measured in 2020. We compared the results initially obtained (i.e., with the distribution of Jumelet et al., 2009) with values retrieved using the same method but assuming another PSD, characterized by a lognormal distribution with a median radius of $1.73\ \mu\text{m}$ and a standard deviation of 1.34 (Deshler et al., 2003).

The optical properties of NAT particles are highly dependent on the size. In Fig. 4, we see that the extinction cross section exhibits a maximum in the ν_2 band of NO_3^- of approximately $19.9\ \mu\text{m}^2$ for the PSD of Deshler et al. (2003), whereas the maximum value is around $13.5\ \mu\text{m}^2$ for Jumelet et al. (2009). Fortunately, as the spectral signature has a similar shape for the 2 PSDs considered, forward simulations for a given OD value result in almost identical spectra, and thereby in similar HRIs and SFs. Accordingly, the LUTs, and the retrieved ODs for measured spectra, are only slightly dependent on the assumed PSD. We compare the LUTs derived from the 2 PSDs considered in Fig. 5a. We also show, as an example, the differences between the forward simulations acquired using the 2 PSDs for a specific case (July, $T_{\text{surf}} = 223\ \text{K}$, Fig. 5b). The retrieval test conducted on IASI-B measurements in 2020 revealed a mean ratio of 0.9 between the ODs retrieved with the PSD of Jumelet et al. (2009) and that of Deshler et al. (2003), which suggests that the uncertainty resulting from the choice of size distribution is on the order of 10% .

The same analysis applies to the shape of the particles, we find the influence of the latter parameter to be negligible (Fig. 4, dashed lines comparing the extinction cross section for spherical and aspherical particles). For the PSD of Jumelet et al. (2009), the maximum extinction cross section is around $12.1\ \mu\text{m}^2$ for prolate spheroids with an axial ratio of 0.5 , whereas the maximum value is $13.5\ \mu\text{m}^2$ for spherical particles.

9 Retrieval of NAT OD over the North Pole

A test was conducted to assess the applicability of the retrieval method to the North Pole. The polar vortex is more unstable above the Arctic, and stratospheric temperatures do not drop to the same low levels as at the South Pole. The greater variability of the Arctic polar vortex implies that PSCs are less frequent and this shows up, for example, in the CALIOP data set, with a factor of 14 between the occurrence frequency of Arctic and Antarctic PSCs (Pitts et al., 2018).

The detection of NAT-containing PSCs from the IASI spectra measured north of $50^\circ\ \text{N}$ has been tested for the months of January, i.e., the month with the highest Arctic PSC occurrence frequency (Pitts et al., 2018). For this, we built a new covariance matrix ($\mathbf{S}$) and a new mean spectrum ($\bar{\mathbf{y}}$), specific for the North Pole. Following the method developed for the South Pole, a total of $900,000$ random IASI-B spectra were selected from those measured north of $50^\circ\ \text{N}$, on two days of each month (except December–February) during the year 2020. The calculation of HRIs for the IASI measurements during the months of January successfully revealed the presence of NAT particles, but over areas and time periods that are, as expected, more limited than those observed at the South Pole. We could confirm the detection of NAT particles by applying the whitening transformation to IASI spectra measured over the North Pole characterized by high HRIs, allowing us to highlight their characteristic spectral signature (see Appendix C).

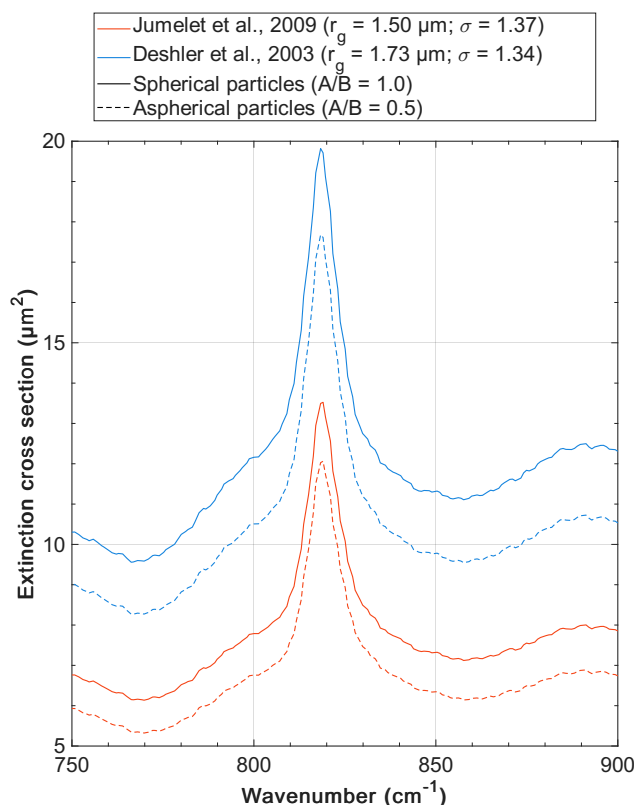


Figure 4. Extinction cross section of NAT particles between 750 and 900 cm^{-1} for 2 different lognormal particle size distributions (Jumelet et al., 2009, in blue, and Deshler et al., 2003, in orange). The solid lines correspond to spherical particles, and the dashed lines to prolate spheroids with an axial ratio (A/B) of 0.5.

Following the correction of the unexpected trends observed in the HRI time series over the North Pole (see Appendix D), we processed the data set in order to obtain NAT optical depths. New forward simulations were done using 4A/OP for the mean profiles of atmospheric temperatures and water vapor over the North Pole in January 2020. The other forward-model parameters were not changed. With this new set of simulated spectra, we built a LUT specific for the North Pole during the month of January, that was then used to convert the HRIs calculated from IASI spectra in ODs.

Figure 6 shows the resulting distributions for six consecutive days in early January 2016 from IASI-B measurements. A major event of NAT particles is observed on 1 January, and lasted for a few days. This result is consistent with an exceptional winter season for the North Pole, which was marked by an unusual period of extremely low temperatures. Pitts et al. (2018) also demonstrated, using CALIOP observations, that the winter of 2015–2016 was an exceptional season for Arctic PSCs.

Note that we retrieve a larger number of negative OD values, compared to the South Pole (Fig. 6 against Fig. 2d). These outliers exceed the expected distribution around 0 due to the HRIs. As they are localized over specific land areas, they are most

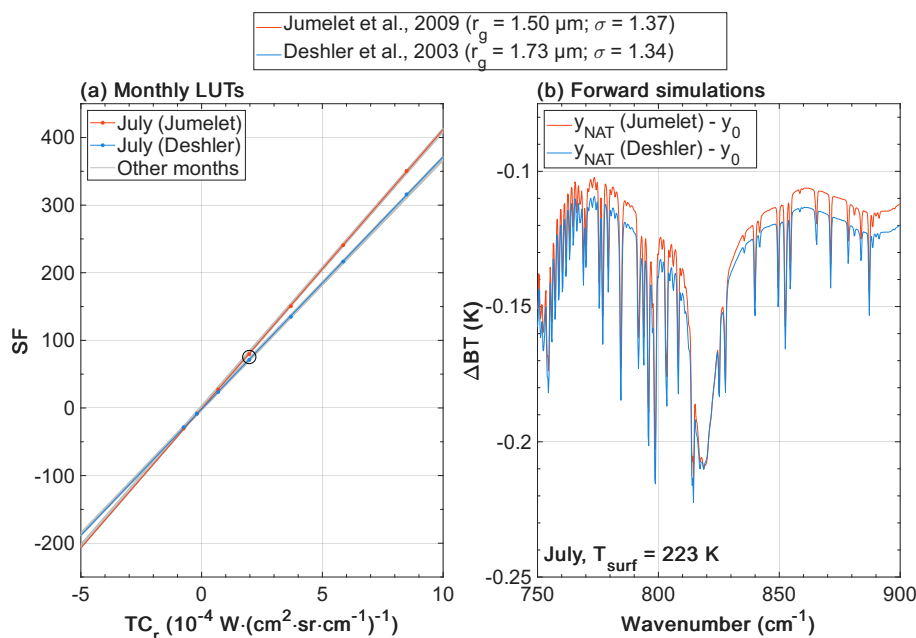


Figure 5. (a) Comparison of the monthly lookup tables (LUTs) obtained for 2 different lognormal particle size distributions (PSDs): in blue, the LUTs based on the PSD of Jumelet et al. (2009) (i.e., the LUTs used in this work to convert NAT HRIs calculated from IASI measurements in ODs), and in orange, the LUTs based on the PSD of Deshler et al. (2003). (b) Illustration of the differences observed between the spectra simulated for the 2 PSDs in a specific case (July, $T_{\text{surf}} = 223 \text{ K}$): the brightness temperature difference between the spectrum simulated with NAT particles ($\text{OD} = 0.01$ at $\lambda = 12.2 \mu\text{m}$) and the equivalent reference spectrum simulated without NAT particles is plotted for each PSD.

likely attributed to a greater variability of the surface emissivity, which is underrepresented in the set of spectra selected for the covariance matrix and the mean spectrum.

Unlike for the South Pole, we do not show monthly distributions of NAT ODs over the North Pole because Arctic NAT-containing PSCs are not frequent enough to be visible when averaged over such extended periods. Nevertheless, it should be noted that the example presented in Fig. 6 is not unique, and that other events with localized high NAT OD values have been identified in the IASI data set.

10 Conclusions

Particles of nitric acid trihydrate (NAT, $\text{HNO}_3 \cdot 3\text{H}_2\text{O}$) are an important constituent of polar stratospheric clouds (PSCs). They constitute therefore an essential element for our understanding of the processes leading to the formation of the Antarctic ozone hole and of more sporadic mini-holes above the Arctic. The first nadir-viewing observations of NAT particles over the South Pole have been reported in a previous work, using the hyperspectral measurements from the Infrared Atmospheric

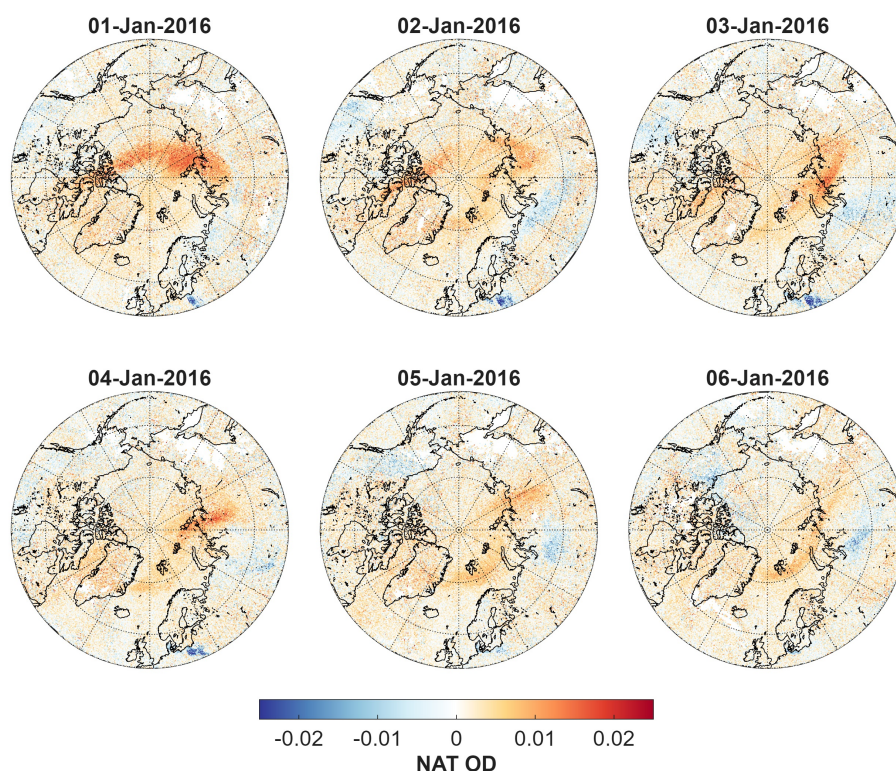


Figure 6. Daily maps of NAT ODs over the North Pole retrieved from IASI-B measurements between 1 and 6 January 2016. The areas with no observations correspond to values excluded during the post-processing.

300 Sounding Interferometer (IASI). This yielded an innovative data set for the detection of NAT-containing PSCs in terms of spatial and temporal sampling. The latter is, however, difficult to exploit due to its simultaneous dependence on the particle column abundance and thermal contrast. In this paper, we have described a retrieval approach relying on the conversion of the detection indices in optical depths of NAT particles (NAT ODs). The lookup tables (LUTs) required for the conversion have been obtained by radiative transfer simulations representing the specific conditions of the Antarctic region, for a set of different thermal contrast situations.

305 The retrieval method has been successfully applied to all IASI measurements south of 50° S, resulting in a coherent record of NAT ODs over a period of more than 18 years (October 2007–December 2025). First distributions of NAT ODs have been presented and compared to the distributions of the initial detection indices. This analysis revealed the efficacy of the retrieval process in addressing the disparities in thermal contrast over the South Pole. Along with the retrieved ODs, we have estimated the uncertainties on a single observation basis, which have been used to exclude clear outliers. The observations associated with the largest uncertainties were particularly found over East Antarctica and over cloudy scenes (i.e., over regions characterized by a weak thermal contrast, and, consequently, a reduced sensitivity). A sensitivity analysis was conducted in order to provide valuable insights into how the assumptions regarding particle size and shape affect the results.



We completed this work by applying the methodology to the North Pole. The detection indexes calculated during the months of January 2008–2025 revealed the sporadic presence of NAT particles. The same approach as for the South Pole was adopted to retrieve NAT ODs using a LUT specifically developed for the month of January at the North Pole, and the results of successful retrievals have been shown for a favorable event in January 2016.

The IASI data set of NAT optical depths constitutes a unique quantitative product for these particles constituting polar stratospheric clouds. We anticipate that this new 18-year record will provide a valuable opportunity to explore the chemistry of the polar stratosphere, when analyzed in conjunction with other variables. Furthermore, the new generation of IASI sounders (IASI-NG), in addition to ensuring the continuity of the measurements, could significantly improve the retrieval of NAT particles, thanks to its improved radiometric performances.

Data availability. The IASI NAT OD data set will be publicly accessible via <https://doi.org/10.5281/zenodo.21835216> (Hermans et al., 2026a) upon publication.

Appendix A: Improving the corrections of the trends in the HRI time series over the South Pole

In our previous work, a global analysis of the HRI time series showed small offsets between the 3 IASI sounders and an unexpected increasing trend of the retrieved values. Corrections have been applied to the initial HRIs to achieve a more consistent data set (Hermans et al., 2026b). By doing so, we assumed that the biases were uniformly distributed in our data set. However, the long-term trends observed in the HRI time series have been found to be dependent on the temperature. We have therefore implemented an additional correction, which is dependent on the baseline temperature of the measured spectra (median value for wavenumbers between 825 and 900 cm^{-1}). For this, we examined daily time series in 6 temperature bins of 10 K and calculated a linear regression based on a limited period of 4 months (December–March). The HRIs associated with each individual measurement were then corrected by interpolation of the linear trends at the corresponding baseline temperature. The impact of the new correction on the HRI time series is illustrated in Figure A1.

Appendix B: South Pole distributions of NAT HRIs

The purpose of this section is to provide a similar figure as for the NAT ODs in Fig. 3, but for the initial detection indices, the NAT HRIs. Figure B1 shows the monthly mean distributions of NAT HRIs on $1^\circ \times 1^\circ$ grid, derived from measurements of IASI-A, B, and C between 2008 and 2025. The comparison between the HRI and OD distributions reveals the effect of the retrieval process, which allows to obtain more homogeneous distributions of NAT particles over the South Pole, by taking the thermal contrast into account.

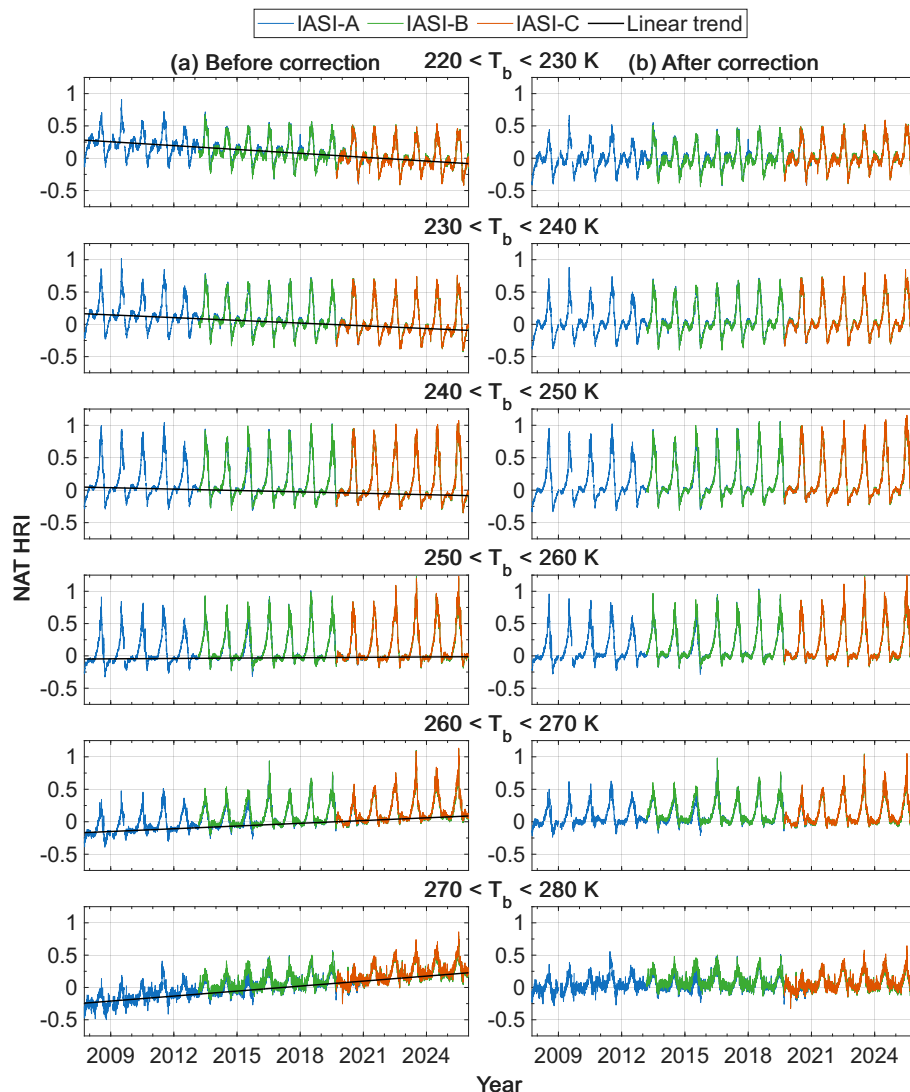


Figure A1. Daily time series of the mean NAT HRIs in 6 different bins of baseline temperature (T_b): (a) after the corrections described in Hermans et al. (2026b), (b) after implementing an additional correction for the residual trends depending on the baseline temperature. The average is calculated based on all measurements south of 50° S for each IASI sounder separately (IASI-A in blue, IASI-B in green, and IASI-C in orange). The linear trends for each T_b bin, plotted in black in panels (a), are derived from a limited period of 4 months (December–March).

Appendix C: Spectral evidence of NAT particles in IASI spectra measured over the North Pole

The whitening transformation was used to confirm the detection of NAT particles in spectra measured over the North Pole associated with high HRIs. This transformation results in a "whitened" spectrum ($\tilde{y}$), in which unusual features are revealed

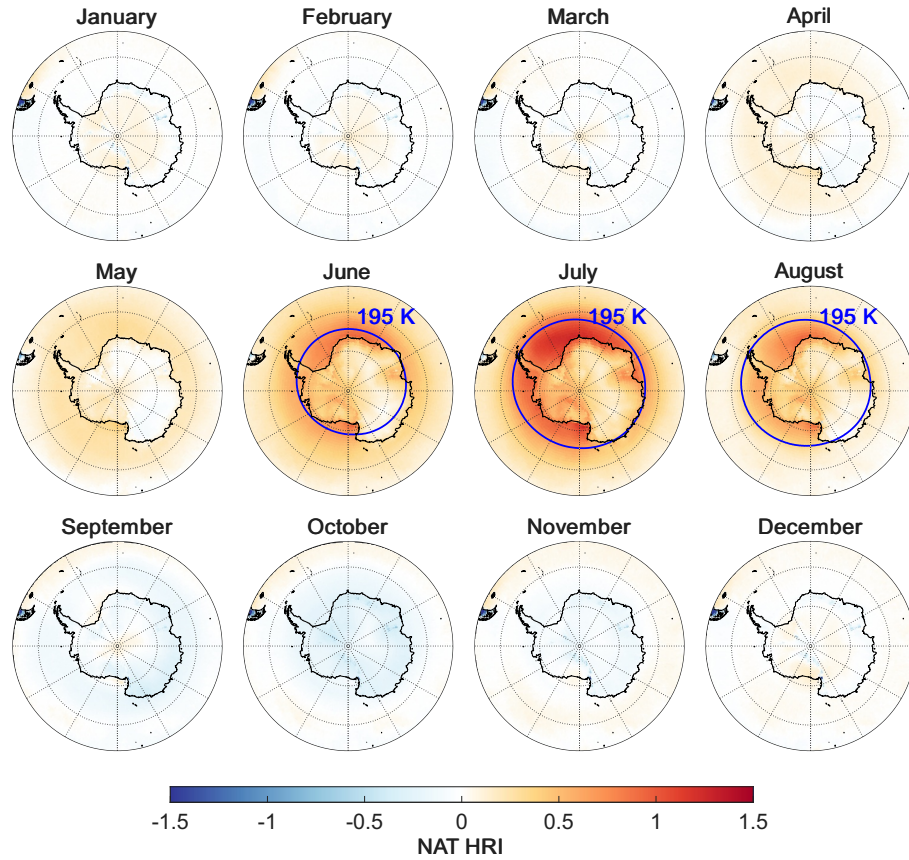


Figure B1. 2008–2025 monthly mean distributions of NAT HRIs on a 1° longitude by 1° latitude grid, derived from measurements of IASI-A, B, and C. The data are first gridded monthly by year, and then an average is calculated based on these annual monthly distributions. The blue contours for the winter months (June–July–August) mark the T_{NAT} value of 195 K at a pressure level of ~ 50 hPa.

by removing the background signal represented by a covariance matrix ($\mathbf{S}$) and a mean spectrum ($\bar{\mathbf{y}}$) (De Longueville et al.,
 345 2021; Franco et al., 2022):

$$\tilde{\mathbf{y}} = \mathbf{S}^{-1/2}(\mathbf{y} - \bar{\mathbf{y}}) \quad (\text{C1})$$

In Fig. C2, we show the result of the whitening transformation applied to a mean spectrum. The average is calculated for IASI-B spectra with an HRI > 3 , measured between 24 and 31 January 2020 in the region revealing the highest HRI values ($0^\circ < \text{longitude} < 60^\circ \text{E}$). The corresponding daily HRI maps are provided in Fig. C1. Superimposed on the whitened spectrum
 350 (in blue in Fig. C2a), the whitened Jacobian of NAT particles ($\tilde{\mathbf{K}}_{\text{NAT}}$, in orange) confirms that the spectral feature observed aligns with the specific NAT signature (attributed to the ν_2 band of NO_3^-). The whitened Jacobian is obtained following:

$$\tilde{\mathbf{K}} = \frac{(\mathbf{S}^{-1/2}\mathbf{K})^T}{\sqrt{\mathbf{K}^T\mathbf{S}^{-1}\mathbf{K}}} \quad (\text{C2})$$

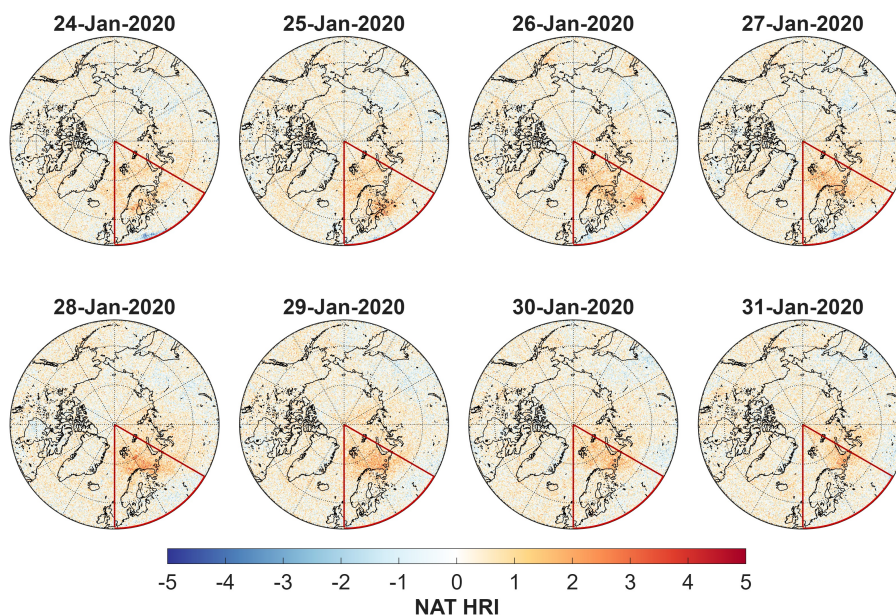


Figure C1. Daily HRI maps over the North Pole, obtained from IASI-B measurements between 24 and 31 January 2020. The red box delimits the region with the highest HRI values, used for the selection of spectra for the whitening transformation illustrated in Fig. C2.

The feature observed in the whitened spectrum around 850 cm^{-1} is likely due to CFC-11 and was already observed in whitened spectra over the South Pole (Hermans et al., 2026b).

355 The concept of the HRI is directly linked to the whitening by:

$$\text{HRI} = \sum_i \text{HRI}_i = \sum_i \tilde{\mathbf{K}}_i \tilde{\mathbf{y}}_i \quad (\text{C3})$$

where the partial HRI, HRI_i , is the contribution of the spectral channel i to the total HRI. This is represented in Fig. C2b and shows that the most significant contributions come from the channels affected by the NAT feature.

Appendix D: Corrections of the trends in the HRI time series over the North Pole

360 As we calculated HRIs only for IASI spectra measured over the North Pole (north of 50° N) during the months of January, we checked for potential unexpected trends in the background values as follows. To consider only the background values, we averaged the HRIs with an absolute value below 1.5 for every January between 2008 and 2025. As a result, our trend analysis is not affected by the measurements with NAT particles, whose presence varies greatly from one year to another. We built time series by plotting the mean values obtained for 7 bins of baseline temperature (between 220 and 290 K) in the middle of every
 365 January (Fig. D1a). We then derived trends by linear fitting, and corrected the HRI data set for the North Pole on a individual basis by interpolating the trends on the measurement date and at the corresponding baseline temperature.

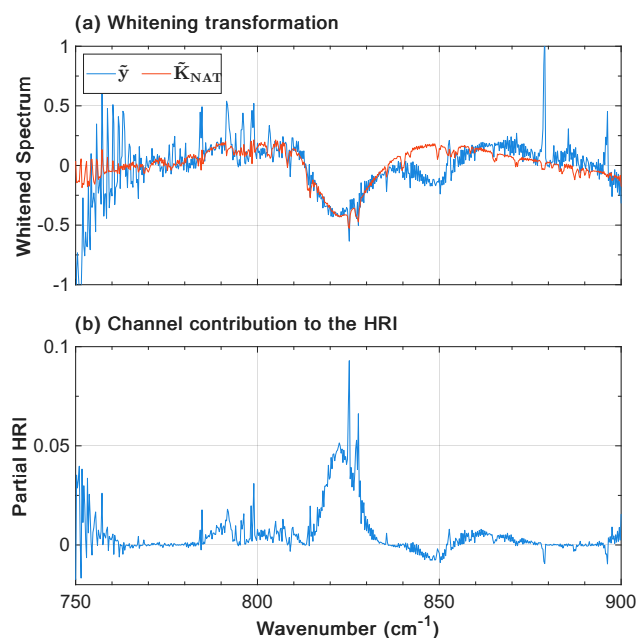


Figure C2. Whitening transformation applied to a mean spectrum calculated for IASI-B spectra, corresponding to an HRI > 3, measured in the region delimited in red in Fig. C1 between 24 and 31 January 2020. (a) In blue, the whitened spectrum ($\tilde{y}$), and, in orange, the whitened Jacobian of NAT particles ($\tilde{K}_{NAT}$), scaled to best match the signature observed in the whitened spectrum. (b) The channel contribution to the HRI.

Author contributions. MH, LC and PC conceptualized the research, developed the retrieval method and analyzed the results. MH prepared the figures and wrote the first version of the manuscript. All authors took part in the discussions and in the revisions of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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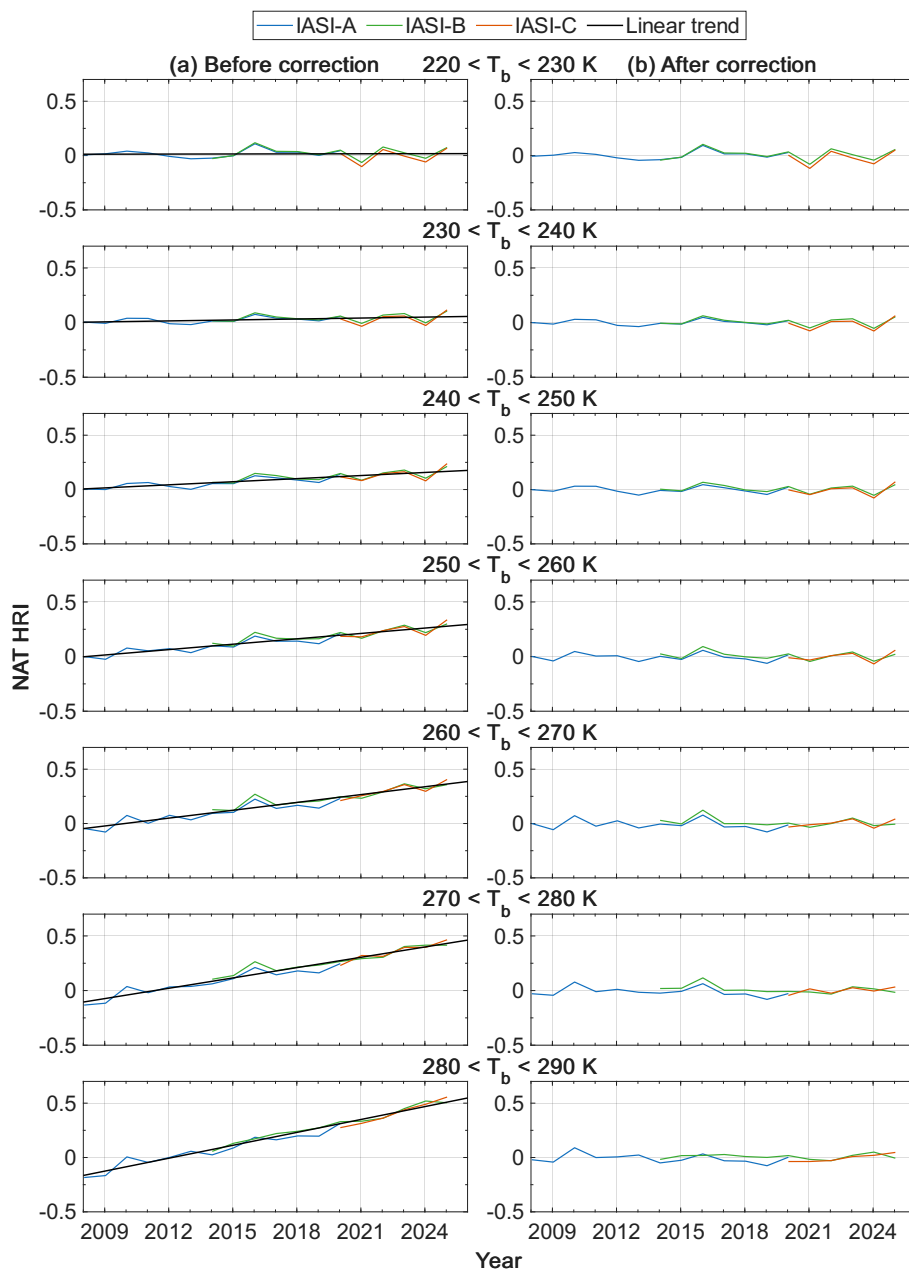


Figure D1. Illustration of the corrections applied to the HRIs calculated over the North Pole. The HRIs obtained during the month of January with an absolute value below 1.5 are averaged by year as a function of the baseline temperature for each IASI sounder separately (IASI-A in blue, IASI-B in green, and IASI-C in orange). The mean values are plotted on 15 January of each year: in panels (a), before correction, with a linear fit of the trends (in black), and in panels (b), after corrections of these trends.



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