Reviewing Global Estimates of Surface Reactive Nitrogen Concentration and Deposition Using Satellite Observation

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Abstract

Since the industrial revolution, human activities have dramatically changed the nitrogen (N) cycle in natural systems. Anthropogenic emissions of reactive nitrogen (N_r) can return to the earth’s surface through atmospheric N_r deposition. Increased N_r deposition may improve ecosystem productivity. However, excessive N_r deposition can cause a series of negative effects on ecosystem health, biodiversity, soil, and water. Thus, accurate estimations of N_r deposition are necessary for evaluating its
environmental impacts. The United States, Canada and Europe have successively launched a number of satellites with sensors that allow retrieval of atmospheric NO\textsubscript{2} and NH\textsubscript{3} column density, and therefore estimation of surface Nr concentration and deposition at an unprecedented spatiotemporal scale. Atmosphere NH\textsubscript{3} column can be retrieved from atmospheric infra-red emission measured by IASI, AIRS, CrIS or TES, while atmospheric NO\textsubscript{2} column can be retrieved from reflected solar radiation measured by GOME, GOME-2, SCIAMACHY, OMI, TEMPO, Sentinel and GEMS. In recent years, scientists attempted to estimate surface Nr concentration and deposition using satellite retrieval of atmospheric NO\textsubscript{2} and NH\textsubscript{3} columns. In this study, we give a thorough review on recent advances of estimating surface Nr concentration and deposition using the satellite retrievals of NO\textsubscript{2} and NH\textsubscript{3}, present a framework of using satellite data to estimate surface Nr concentration and deposition based on recent works, and summarize the existing challenges for estimating surface Nr concentration and deposition using the satellite-based methods. We believe that exploiting satellite data to estimate Nr deposition has a broad and promising prospect.

**Keywords**
Nitrogen deposition; Satellite retrieval; Surface concentration; Oxidized and reduced Nr

**1. Introduction**
Nitrogen (N) exists in three forms in the environment including reactive nitrogen (Nr), organic nitrogen (ON) and nitrogen gas (N\textsubscript{2}) (Canfield et al., 2010). N\textsubscript{2} is the main component of air, accounting for 78% of the total volume of air, but it cannot be directly used by most plants. Nr (such as NO\textsubscript{3}\textsuperscript{-} and NH\textsubscript{4}\textsuperscript{+}) is the main form of N that can be directly used by most plants, but the content of Nr in nature is much lower compared with ON and N\textsubscript{2} (Vitousek et al., 1997; Nicolas and Galloway, 2008).
supply of N_r is essential for all life forms and contributes to the increase in
government production, thus providing sufficient food for the growing global
population (Galloway et al., 2008; David et al., 2013; Galloway et al., 2004b; Erisman
et al., 2008). Before the industrial revolution, N_r mainly came from natural sources
such as biological N fixation, lightning and volcanic eruption (Galloway et al., 2004a).
Since the industrial revolution, human activities (e.g. agricultural development,
combustion of mineral energy) have greatly perturbed the N cycle in natural systems
(Canfield et al., 2010; Kim et al., 2014; Lamarque et al., 2005).

N_r (NO_x and NH_3) emitted to the atmosphere will return to the earth surface through
atmospheric deposition (Liu et al., 2011). Atmospheric N_r deposition refers to the
process in which N_r are removed from the atmosphere, including wet (rain and snow)
and dry (gravitational settling, atmospheric turbulence, etc.) deposition (Xu et al.,
2015; Zhang et al., 2012; Pan et al., 2012). The input of N_r over terrestrial natural
ecosystems primarily comes from the N_r deposition (Shen et al., 2013; Sutton et al.,
2001; Larsen et al., 2011). In the short term, atmospheric N_r deposition can increase
the N_input to ecosystems, which promotes plant growth and enhances ecosystem
productivity (Erisman et al., 2008; Sutton et al., 2013). However, excessive
atmospheric N_r deposition also causes a series of environmental problems (Liu et al.,
2017d). Due to the low efficiency of agricultural N application, plenty of N_r is lost
through runoff, leaching and volatilization, causing serious environmental pollution.
Excessive N_r deposition may aggravate the plant’s susceptibility to drought or frost,
reduce the resistance of plant to pathogens or pests, and further affect the physiology
and biomass distribution of vegetation (ratio of roots, stems and leaves) (Stevens et al.,
2004; Nadelhofer et al., 1999; Bobbink et al., 2010; Janssens et al., 2010). Excessive
N_r leads to eutrophication and related algal blooms over aquatic ecosystems, reducing
water biodiversity (Paerl et al., 2014), while excessive N$_r$ in drinking water also poses a threat to human health (Zhao et al., 2013). Therefore, monitoring and estimation of surface N$_r$ concentration and deposition on the global scale are of great importance and urgency.

The methods of estimating atmospheric N$_r$ deposition can be divided into three categories: ground-based monitoring, atmospheric chemical transport modeling (ACTM) and satellite-based estimation. Ground-based monitoring is considered to be the most accurate quantitative method, which can effectively reflect the N$_r$ deposition in local areas. ACTM can simulate the processes of N$_r$ chemical reaction, transport, and deposition, as well as the vertical distribution of N$_r$. Satellite-based estimation establishes empirical, physical or semi-empirical models by connecting the ground-based N$_r$ concentrations and deposition with satellite-derived N$_r$ concentration. This study focuses on reviewing the recent development of satellite-based methods to estimate N$_r$ deposition. We firstly give a brief introduction to the progress of ground-based monitoring, ACTM-based methods, and then present a detailed framework of using satellite observation to estimate dry and wet N$_r$ deposition (including both oxidized and reduced N$_r$). Next, we review the recent advances of the satellite-based methods of estimating N$_r$ deposition. Finally, we discuss the remaining challenges for estimating surface N$_r$ concentration and deposition using satellite observation.

2.1 Methods for Estimating Surface N$_r$ Concentration and Deposition

2.1.1 Ground-based Monitoring

Ground-based monitoring of N$_r$ deposition can be divided into two parts: wet and dry N$_r$ deposition monitoring. Since the 1970s, there have been large-scale monitoring networks focusing on the wet N$_r$ deposition. The main large-scale regional monitoring
networks include Canadian Air and Precipitation Monitoring Network (CAPMoN), Acid Deposition Monitoring Network in East Asia (EANET), European Monitoring and Evaluation Program (EMEP), United States National Atmospheric Deposition Program (NADP), World Meteorological Organization Global Atmosphere Watch Precipitation Chemistry Program, and Nationwide Nitrogen Deposition Monitoring Network in China (NNDMN) (Tan et al., 2018; Vet et al., 2014). The detailed scientific objectives of the wet Nr deposition observation networks vary, but most of the observation networks mainly concentrate on the spatiotemporal variation of wet deposition of ions including Nr compounds, the long-term trends of ions in precipitation, and the evaluation of ACTMs.

Compared with wet Nr deposition monitoring, dry Nr deposition monitoring started late, due to the limitation of monitoring technology since it is more difficult to be quantified (affected greatly by surface roughness, air humidity, climate and other environmental factors) (Liu et al., 2017c). Dry Nr deposition observation networks include US ammonia monitoring network (AMoN), CAPMoN, EANET and EMEP. The monitoring methods of dry Nr deposition are mainly divided into direct monitoring (such as dynamic chambers) and indirect monitoring (such as inferential methods). The inferential model is widely applied in ground-based monitoring networks (such as EANET and NNDMN), mainly because this method is more practical and simpler. In inferential models, dry deposition is divided into two parts: surface Nr concentrations and the deposition velocity \( V_d \) of Nr (Nowlan et al., 2014). \( V_d \) can be estimated by meteorology, land use types of underlying surface as well as the characteristics of each Nr component itself using resistance models (Nemitz et al., 2001). Thus, dry Nr deposition monitoring networks only need to focus on the quantification of surface concentration of individual Nr components. The Nr
components in the atmosphere are very complex, including N$_2$O$_5$, HONO, NH$_3$, NO$_2$, HNO$_3$ and particulate NH$_4^+$ and NO$_3^-$. Most monitoring networks include the major N$_r$ species such as gaseous NH$_3$, NO$_2$, HNO$_3$ and the particles of NH$_4^+$ and NO$_3^-$. Effort of ground-based N$_r$ deposition monitoring mostly concentrates on wet N$_r$ deposition, while observations of dry N$_r$ deposition are relatively scarce especially for surface HNO$_3$ and NH$_4^+$ and NO$_3^-$. Second, most observation networks focus on a few years or a certain period of time, leading to the lack of long-term continuously monitoring on both wet and dry N$_r$ deposition. More importantly, the global N$_r$ deposition monitoring network has not been established, and the sampling standards in different regions are not unified. These outline the potential room for improvement of ground-based N$_r$ deposition monitoring.

2.1.2 ACTM Simulation

An ACTM can simulate N$_r$ deposition at regional or global scales through explicitly representing the physical and chemical processes of atmospheric N$_r$ components (Zhao et al., 2017; Zhang et al., 2012). Wet N$_r$ deposition flux is parameterized as in-cloud, under-cloud and precipitation scavenging (Amos et al., 2012; Levine and Schwartz, 1982; Liu et al., 2001; Mari et al., 2000), while dry deposition flux can be obtained as the product of surface N$_r$ concentration and $V_d$, which is typically parameterized as a network of resistances (Wesely and Hicks, 1977). Based on the integrated results of 11 models of HTAP (hemispheric transport of air pollution), Tian et al. found that about 76%-83% of the ACTM’s simulation results were ±50% of the monitoring values, and the modeling results underestimated the wet deposition of NH$_4^+$ and NO$_3^-$ over Europe and East Asia, and overestimated the wet deposition of NO$_3^-$ over the eastern US. Though regional ACTMs can be configured at very high horizontal resolution (e.g., 1×1 km$^2$) (Kuik et al., 2016), the horizontal resolution of
global ACTMs are relatively coarse (1° × 1°-5° × 4°) (Williams et al., 2017), which cannot indicate the local pattern of N\textsubscript{r} deposition. On the other hand, the N\textsubscript{r} emission inventory used to drive an ACTM is highly uncertain, with the uncertainty of the NO\textsubscript{x} emission at about ±30-40%, and that of NH\textsubscript{3} emission at about ±30-80% (Zhang et al., 2009; Cao et al., 2011).

### 2.1.3 Satellite-based Estimation of Surface N\textsubscript{r} Concentration and Deposition

Satellite observation has wide spatial coverages and high resolution, and is spatiotemporally continuous. Atmospheric NO\textsubscript{2} and NH\textsubscript{3} columns can be derived from satellite measurements with relatively high accuracy (Van Damme et al., 2014a; Boersma et al., 2011), providing a new perspective about atmospheric N\textsubscript{r} abundance.

Satellite instruments that can monitor NO\textsubscript{2} in the atmosphere include GOME (Global Ozone Monitoring Experience), SCIAMACHY (SCanning Imaging Absorption SpectroMeter for Atmospheric ChartographY), OMI (Ozone Monitoring Instrument), GOME-2 (Global Ozone Monitoring Experience-2). Some scholars applied satellite NO\textsubscript{2} columns to estimate the surface NO\textsubscript{2} concentration, and then dry NO\textsubscript{2} deposition by combining the surface NO\textsubscript{2} concentration and modeled V\textsubscript{d}. Cheng et al. (Cheng et al., 2013) established a statistical model to estimate the surface NO\textsubscript{2} concentration based on the SCIAMACHY NO\textsubscript{2} columns, and then estimated the dry deposition of NO\textsubscript{2} over eastern China. This method by Cheng et al. (Cheng et al., 2013) using the simple linear model did not consider the vertical profiles of NO\textsubscript{2}. Lu et al. (Lu et al., 2013) established a multivariate linear regression model based on the SCIAMACHY and GOME NO\textsubscript{2} columns, meteorological data and ground-based monitoring N\textsubscript{r} deposition, and then estimated the global total N\textsubscript{r} deposition. Lu et al. (Lu et al., 2013) could not distinguish the contribution of dry and wet N\textsubscript{r} deposition using the
multivariate linear regression model. Jia et al. (Jia et al., 2016) established a simple linear regression model based on OMI tropospheric NO$_2$ column and ground-based surface N$_r$ concentration, and then estimated the total amounts of dry N$_r$ deposition. Jia et al. (Jia et al., 2016) used OMI tropospheric NO$_2$ column to estimate the dry deposition of reduced N$_r$ deposition (NH$_3$ and NH$_4^+$), which could also bring great errors since the OMI NO$_2$ column could not indicate the NH$_3$ emission. These studies highlight the problem of using only NO$_2$ columns to derive total N$_r$ deposition, that NO$_2$ columns give us highly limited information about the abundance of reduced N$_r$ (NH$_3$ and NH$_4^+$).

Lamsal et al. (Lamsal et al., 2008) first used the relationship between the NO$_2$ column and surface NO$_2$ concentration at the bottom layer simulated by an ACTM to convert OMI NO$_2$ column to surface NO$_2$ concentration. A series of works (Lamsal et al., 2013; Nowlan et al., 2014; Kharol et al., 2018) have effectively estimated regional and global surface NO$_2$ concentration using satellite NO$_2$ column combining with ACTM-derived relationship between the NO$_2$ column and surface NO$_2$ concentration simulated. It is worth mentioning that Nowlan et al. (Nowlan et al., 2014) applied OMI NO$_2$ column to obtain the global dry NO$_2$ deposition during 2005-2007 for the first time. However, using satellite NO$_2$ column and ACTM-derived relationship between the NO$_2$ column and surface NO$_2$ concentration may lead to an underestimation of surface NO$_2$ concentration. Kharol et al. (Kharol et al., 2015) found that the satellite-derived surface NO$_2$ concentration using the above method is only half of the observed values. To resolve such potential underestimation, Larkin et al. (Larkin et al., 2017) established a statistical relationship between the satellite-derived and ground measured surface NO$_2$ concentration, and then calibrated the satellite-derived surface NO$_2$ concentration using the established relationship.
Some researchers also estimated other N\textsubscript{r} components (such as particulate NO\textsubscript{3}\textsuperscript{-}) based on satellite NO\textsubscript{2} column. Based on the linear model between NO\textsubscript{2}, NO\textsubscript{3}\textsuperscript{-}, HNO\textsubscript{3} obtained by ground-based measurements, Jia et al. (Jia et al., 2016) calculated the surface NO\textsubscript{3}\textsuperscript{-} and HNO\textsubscript{3} concentration using satellite-derived surface NO\textsubscript{2} concentration and their relationship. Geddes et al. (Geddes and Martin, 2017) reconstructed the NO\textsubscript{x} emission data by using the satellite NO\textsubscript{2} column, and then estimated the global NO\textsubscript{x} deposition by an ACTM, but the spatial resolution of global NO\textsubscript{x} deposition remains low (2°x2.5°), failing to exploit the higher resolution of satellite observation.

Comparing with NO\textsubscript{2}, the development of satellite NH\textsubscript{3} monitoring is relatively late. Atmospheric NH\textsubscript{3} was first detected by the TES in Beijing and Los Angeles (Beer et al., 2008). The IASI sensor also detected atmospheric NH\textsubscript{3} from a biomass burning event in Greece (Coheur et al., 2009). Subsequently, many scholars began to develop more reliable satellite NH\textsubscript{3} column retrievals (Whitburn et al., 2016; Van Damme et al., 2014a), validate the satellite-retrieved NH\textsubscript{3} column with the ground-based observation (Van Damme et al., 2014a; Dammers et al., 2016; Li et al., 2017), and compare the satellite NH\textsubscript{3} column with the aircraft measured NH\textsubscript{3} column (Van Damme et al., 2014b; Whitburn et al., 2016). In recent years, some scholars have carried out the works of estimating surface NH\textsubscript{3} concentration based on satellite NH\textsubscript{3} column. Liu et al. (Liu et al., 2017b) obtained the satellite-derived surface NH\textsubscript{3} concentration in China based on the IASI NH\textsubscript{3} column coupled with an ACTM, and deepened the understanding of the spatial pattern of surface NH\textsubscript{3} concentration in China. Similarly, Graaf et al. (Van der Graaf et al., 2018) carried out the relevant work in Europe based on the IASI NH\textsubscript{3} column coupled with an ACTM, and estimated the dry NH\textsubscript{3} deposition in West Europe. Jia et al. (Jia et al., 2016) first constructed the linear model
between surface NO$_2$ and NH$_4^+$ concentration based on ground monitoring data, and then calculated the NH$_4^+$ concentration using satellite-derived surface NO$_2$ concentration and their relationship. However, as the emission sources of NO$_x$ (mainly from transportation and energy sectors) and NH$_3$ (mainly from agricultural sector) are different (Hoesly et al., 2018), the linear model between surface NO$_2$ and NH$_4^+$ concentration may lead to large uncertainties in estimating the global NH$_4^+$ concentration. There is still no report about the satellite-derived dry and wet reduced N$_r$ deposition using satellite NH$_3$ column at a global scale. As reduced N$_r$ plays an important role in total N$_r$ deposition, satellite NH$_3$ should be better utilized to help estimate reduced N$_r$ deposition.

### 2.1.4 Problems in Estimating Global N$_r$ Deposition

The spatial coverage of ground monitoring sites focusing on N$_r$ deposition is still not adequate, and the monitoring standards and specifications in different regions of the world are not consistent, presenting a barrier to integrating different regional monitoring data. Large uncertainties exist in N$_r$ emission inventory used to drive the ACTMs, and the spatial resolution of the modeled N$_r$ deposition by ACTMs is coarse. Using satellite monitoring data to estimate surface N$_r$ concentration and deposition is still in its infancy, especially for reduced N$_r$.

Some scholars tried to use satellite NO$_2$ and NH$_3$ column to estimate the surface N$_r$ concentration and dry N$_r$ deposition. However, there are relatively few studies on estimating wet N$_r$ deposition. In addition, the development of satellite monitoring for NH$_3$ in the atmosphere is relatively late (compared with NO$_2$). At present, IASI NH$_3$ data have been widely used, while the effective measurements of TES are less than IASI; CrIS and AIRS NH$_3$ column products are still under development. There are three main concerns in high-resolution estimation of surface N$_r$ concentration and
deposition based on satellite Nr observation. (1) How to effectively couple the satellite high-resolution NO$_2$ and NH$_3$ column data with the vertical profiles simulated by an ACTM, and then estimates the surface Nr concentrations? This step is the key to simulate the dry Nr deposition. (2) How to construct a model for estimating dry Nr deposition including all major Nr species based on satellite NO$_2$ and NH$_3$ column, and then estimates the dry Nr deposition at a high spatial resolution? (3) How to combine the high-resolution satellite NO$_2$ and NH$_3$ column data and ground-based monitoring data to construct wet Nr deposition models, and then estimate the wet Nr deposition at a high spatial resolution?

3. Framework of Estimating Surface Nr Concentration and Deposition Using Satellite Observation

We give a framework of using satellite observation to estimate surface Nr concentration and deposition as shown in Fig. 1 based on recent advances.

Fig. 1 Schematic diagram of dry and wet Nr deposition. (a) indicates satellite observed NO$_2$ and NH$_3$ column, and the vertical profiles by an ACTM; (b) shows dry and wet Nr deposition including the major Nr species (gaseous NO$_2$, HNO$_3$, NH$_3$, particulate NO$_3^-$ and NH$_4^+$, as well as wet NO$_3^-$ and NH$_4^+$ in precipitation); (c) illustrates atmospheric vertical structures including the troposphere (satellite observation), atmospheric boundary layer (ABL), interfacial sub-layer; (d) and (e) represent procedures of calculating the dry and wet Nr deposition.

3.1.1 Conversion of Satellite NO$_2$ and NH$_3$ Column to Surface Nr Concentration

An ACTM can simulate the vertical profiles of NO$_2$ and NH$_3$ with multiple layers
from the surface to the troposphere. For example, the GEOS-Chem ACTM includes 274 vertical layers from the earth surface to the top of the stratosphere. Most previous studies estimated the ratio of surface \( N_r \) concentration (at the first layer) to total columns by an ACTM, and then multiply the ratio by satellite columns to estimate satellite-derived surface concentration (Geddes et al., 2016; Graaf et al., 2018; Nowlan et al., 2014).

Another approach tries to fit general vertical profiles of \( \text{NO}_2 \) and \( \text{NH}_3 \) (Zhang et al., 2017; Liu et al., 2017b; Liu et al., 2017c), and then estimate the ratio of \( N_r \) concentration at any height to total \( N_r \) columns, and finally multiply the ratio by satellite \( \text{NO}_2 \) and \( \text{NH}_3 \) columns. This approach has an advantage compared with the previous one for that \( \text{NO}_2 \) and \( \text{NH}_3 \) concentration at all altitude included in ACTM simulations can be estimated.

Taking the estimation of surface \( \text{NO}_2 \) concentration using the latter approach as an example, the methods and steps are introduced in the following.

Step 1: Calculate the monthly mean \( \text{NO}_2 \) concentrations at all layers simulated by an ACTM.

Step 2: Construct the vertical profile function of \( \text{NO}_2 \). Multiple Gaussian functions are used to fit the vertical distribution of \( \text{NO}_2 \) based on the monthly \( \text{NO}_2 \) concentrations at all layers calculated in Step 1, in which the independent variable is the height (altitude), and the dependent variable is \( \text{NO}_2 \) concentration at a certain height.

The basic form of single Gaussian function is (Zhang et al., 2017; Liu et al., 2017b; Liu et al., 2017c; Whitburn et al., 2016):

\[
\rho = \rho_{\text{max}} e^{-\frac{(Z-Z_0)^2}{\sigma^2}} \tag{1}
\]

where \( Z \) is the height of a layer in the ACTM; \( \rho_{\text{max}} \), \( Z_0 \) and \( \sigma \) are the maximum \( \text{NO}_2 \) concentration, the corresponding height with the maximum \( \text{NO}_2 \) concentration and the
There are two basic forms of profile shapes of NO\(_2\): (1) NO\(_2\) concentration reaches the maximum concentration when reaching a certain height (Z\(_0\) ≠ 0). As the height increases, the NO\(_2\) concentration begins to decline; (2) NO\(_2\) concentration is basically concentrated on the earth surface (Z\(_0\) = 0). These two cases are the ideal state of the vertical distribution of NO\(_2\) concentration. In reality, single Gaussian fitting may not capture the vertical distribution of NO\(_2\) well. To improve the accuracy of fitting, the sum of multiple Gaussian functions can be used:

\[
\rho(Z) = \sum_{i=1}^{n} \rho_{\text{max},i} e^{-\frac{(Z-Z_{0,i})^2}{\sigma_i^2}} \tag{2}
\]

Step 3: Calculate the ratio of NO\(_2\) concentration at the height of h\(_G\) to total columns \((\int_0^{h_{\text{trop}}} \rho(Z) \, dx)\), and then multiply the ratio by satellite column (S\(_{\text{trop}}\)). The satellite-derived N\(_r\) concentration at the height of h\(_G\) can be calculated as:

\[
S_{G,\text{NO}_2} = S_{\text{trop}} \times \frac{\rho(h_G)}{\int_0^{h_{\text{trop}}} \rho(Z) \, dx} \tag{3}
\]

Step 4: Convert the instantaneous satellite-derived surface NO\(_2\) concentration \((S_{G,\text{NO}_2} \ast)\) using the ratio of average surface NO\(_2\) concentration \((G_{\text{ACTM}}^{1-2.4})\) to that at satellite overpass time \((G_{\text{ACTM}}^{\text{overpass}}))\) by an ACTM:

\[
S_{G,\text{NO}_2} \ast = \frac{G_{\text{ACTM}}^{1-2.4}}{G_{\text{ACTM}}^{\text{overpass}}} \times S_{G,\text{NO}_2} \tag{4}
\]

The method for estimating the surface NH\(_3\) concentration \((S_{G,\text{NH}_3} \ast)\) is similar to that for estimating the surface NO\(_2\) concentration.

### 3.1.2 Estimating Surface Concentration of Other N\(_r\) Species

At present, only NO\(_2\) and NH\(_3\) column can be retrieved reliably, and there are no reliable satellite retrievals of HNO\(_3\), NH\(_4^+\) and NO\(_3^-\). For example, the IASI HNO\(_3\) product is still in the stage of data development and verification (Ronsmans et al.,...
14. Previous studies firstly derive the relationship between \( N_r \) species by an ACTM or by ground-based measurements, and then use the relationship to convert satellite-derived surface \( \text{NO}_2 \) and \( \text{NH}_3 \) concentration (\( S_{\text{G,NH}_3} \)) to \( \text{HNO}_3 \), \( \text{NH}_4^+ \) and \( \text{NO}_3^- \) concentrations:

\[
\begin{align*}
G_{\text{S,NO}_3} &= S_{\text{G,NO}_2} \times \frac{G_{\text{ACTM,NO}_3}}{G_{\text{ACTM,NO}_2}} \\
G_{\text{S,HNO}_3} &= S_{\text{G,NO}_2} \times \frac{G_{\text{ACTM,HNO}_3}}{G_{\text{ACTM,NO}_2}} \\
G_{\text{S,NH}_4} &= S_{\text{G,NH}_3} \times \frac{G_{\text{ACTM,NH}_4}}{G_{\text{ACTM,NH}_3}}
\end{align*}
\]

\( \frac{G_{\text{ACTM,NO}_3}}{G_{\text{ACTM,NO}_2}} \), \( \frac{G_{\text{ACTM,HNO}_3}}{G_{\text{ACTM,NO}_2}} \), \( \frac{G_{\text{ACTM,NH}_4}}{G_{\text{ACTM,NH}_3}} \) is the estimated ratio of between \( \text{NO}_2 \) and \( \text{NO}_3^- \), \( \text{NO}_2 \) and \( \text{HNO}_3 \), \( \text{NH}_3 \) and \( \text{NH}_4^+ \).

3.1.3 Dry Deposition of \( N_r \)

The resistance of dry \( N_r \) deposition mainly comes from three aspects: aerodynamic resistance (\( R_a \)), quasi laminar sub-layer resistance (\( R_b \)) and canopy resistance (\( R_c \)).

The \( V_d \) can be expressed as

\[
V_d = \frac{1}{R_a + R_b + R_c} + v_g \quad (6)
\]

\( v_g \) is gravitational settling velocity. For gases, the \( v_g \) is negligible (\( v_g \approx 0 \)).

Dry \( \text{NO}_2 \), \( \text{NO}_3^- \), \( \text{HNO}_3 \), and \( \text{NH}_4^+ \) deposition can be calculated by:

\[
F = G_S \times V_d \quad (7)
\]

Unlike above species, \( \text{NH}_3 \) is bi-directional, presenting both upward and downward fluxes. There is a so-called “canopy compensation point” (\( C_o \)) controlling dry \( \text{NH}_3 \) deposition. Dry \( \text{NH}_3 \) deposition can be calculated by:

\[
F = (G_{\text{S,NH}_3} - C_o) \times V_d \quad (8)
\]

The calculation of \( C_o \) is very complex including the leaf stomatal and soil emission potentials related to the meteorological factors, the plant growth stage and the canopy type. The satellite-based methods usually neglected this complex process and set \( C_o \).
as zero (Graaf et al., 2018; Kharol et al., 2018) or set fixed values in each land use type based on ground-based measurements (Jia et al., 2016).

3.1.4 Wet Deposition of $N_r$

The satellite-based estimation of wet $N_r$ deposition can be simplified as the product of the concentration of $N_r$ ($C$), precipitation ($P$) and scavenging coefficient ($w$) (Pan et al., 2012). Satellite NO$_2$ and NH$_3$ can be used to indicate the oxidized $N_r$ and reduced $N_r$; precipitation ($P$) can be obtained from ground monitoring data or reanalysis data (such as NCEP). However, the scavenging coefficient ($w$) is usually highly uncertain. To improve the accuracy of estimation, a mixed-effects model (Liu et al., 2017a; Zhang et al., 2018) is proposed to build the relationship between satellite NO$_2$ and NH$_3$, precipitation and ground monitoring wet $N_r$ deposition:

$$W_{ij} = \alpha_j + \beta_i \times P_{ij} \times (S_{ABL})_{ij} + \epsilon_{ij} \quad (9)$$

$$S_{ABL} = S_{trop} \times \int_0^{ABL} \frac{\rho(Z) \, dz}{\int_0^{trop} \rho(Z) \, dz} \quad (10)$$

$W_{ij}$ is wet NO$_3^-$N or NH$_4^+$-N deposition at month $i$ and site $j$; $(S_{ABL})_{ij}$ is the atmospheric boundary layer (ABL) NO$_2$ or NH$_3$ columns at month $i$ and site $j$; $P_{ij}$ is precipitation at month $i$ and site $j$; $\beta_i$ and $\alpha_j$ are the slope and intercept of random effects, representing seasonal variability and spatial effects; $\epsilon_{ij}$ represents the random error at month $i$ and site $j$.

The scavenging process of wet $N_r$ deposition usually starts from the height of rainfall rather than the top of the troposphere, so it is more reasonable to use NO$_2$ and NH$_3$ column below the height of rainfall to build the wet $N_r$ deposition model. The NO$_2$ and NH$_3$ column within ABL is used to build the wet deposition model since precipitation height is close to the height of the ABL (generally less than 2-3 km).
4. Satellite-derived Surface $N_r$ Concentration and Deposition

4.1 Surface NO$_2$ Concentration and Oxidized $N_r$ Deposition

The spatial resolutions of global ACTMs and therefore modeled surface $N_r$ concentration are very coarse (for example, the spatial resolution of the global version of GEOS-Chem is $2^\circ \times 2.5^\circ$). Thus it can be hard to estimate surface $N_r$ concentration and deposition at a fine resolution at a global scale by ACTMs alone. Instead, the satellite $N_r$ retrievals have a high spatial resolution and can reveal more spatial details than ACTM simulations.

Cheng et al. (Cheng et al., 2013) and Jia et al. (Jia et al., 2016) established a linear model between the surface NO$_2$ concentration and NO$_2$ column by assuming the ratio of the surface NO$_2$ concentration to the tropospheric NO$_2$ column to be fixed, and then used the linear model to convert satellite NO$_2$ columns to surface NO$_2$ concentration, and finally estimated dry NO$_2$ deposition using the inferential method. However, these statistical methods by Cheng et al. (Cheng et al., 2013) and Jia et al. (Jia et al., 2016) are highly dependent on the ground-based measurements, and the established linear models may be not effective over regions with few monitoring sites.

A comprehensive study (Nowlan et al., 2014) estimated global surface NO$_2$ concentration during 2005-2007 by multiplying OMI tropospheric NO$_2$ columns by the ACTM-modeled ratio between surface NO$_2$ concentration and tropospheric column (Fig. 2). Nowlan et al. (Nowlan et al., 2014) also estimated dry NO$_2$ deposition using the OMI-derived surface NO$_2$ concentration combining the modeled $V_d$ during 2005-2007. This approach followed an earlier study (Lamsal et al., 2008), that focus on North America. As reported by Lamsal et al. (Lamsal et al., 2008), the satellite-derived surface NO$_2$ concentration was generally lower than ground-based NO$_2$ observations, ranging from -17% to -36% in North America. Kharol et al.
(Kharol et al., 2015) used a similar method and found the satellite-derived surface NO₂ concentration was only half of the ground-measured values in North America (Kharol et al., 2015).

![Satellite-derived surface NO₂ concentration during 2005-2007 by Nowlan et al. (Nowlan et al., 2014) (a) and by Geddes et al. (Geddes et al., 2016) (b).](image1)

Geddes et al. (Geddes et al., 2016) followed previous studies, and used NO₂ column from the GOME, SCIAMACHY, and GOME-2 to estimate surface NO₂ concentration. Although Geddes et al. (Geddes et al., 2016) did not evaluate their results with ground-based observation, it is obvious that their surface NO₂ estimates were higher than Nowlan’s estimates (Nowlan et al., 2014) based on OMI (Fig. 2). This may be because the OMI-derived NO₂ column is much lower than that derived by GOME, SCIAMACHY, and GOME-2, especially over polluted regions. For example, in China, the OMI NO₂ column is about 30% lower than that of SCIAMACHY and GOME-2 consistently (Fig. 3).

![An example of the time series of monthly NO₂ column retrieved by GOME, SCIAMACHY, GOME2 and OMI in China.](image2)

Larkin et al. (Larkin et al., 2017) established a land-use regression model to estimate
global surface NO$_2$ concentration by combining satellite-derived surface NO$_2$
concentration by Geddes et al. (Geddes et al., 2016) and ground-based annual NO$_2$
measurements. The study by Larkin et al. (Larkin et al., 2017) can be considered as
using the ground-based annual measurements to adjust the satellite-derived surface
NO$_2$ concentration by Geddes et al. (Geddes et al., 2016), which helped reduce the
discrepancy between satellite-derived and ground-measured NO$_2$ concentration. The
regression model captured 54% of global NO$_2$ variation, with an absolute error of 2.32
μg N m$^{-3}$.

Zhang et al. (Zhang et al., 2017) followed the framework in Sect. 3 to estimate the
OMI-derived surface NO$_2$ concentration (at ~50 m) in China, and found good
agreement with ground-based surface NO$_2$ concentration from the NNDMN at yearly
scale (slope=1.00, $R^2$=0.89). The methods by Zhang et al. (Zhang et al., 2017) can
also generate OMI-derived NO$_2$ concentration at any height by the constructed NO$_2$
vertical profile. Zhang et al. (Zhang et al., 2017) also estimated dry NO$_2$ deposition
using the OMI-derived surface NO$_2$ concentration combining the modeled $V_d$ during
2005-2016. Based on Zhang’s estimates, the Gaussian function can well simulate the
vertical distribution of NO$_2$ from an ACTM (MOZART) (Emmons et al., 2010) with
99.64% of the grids having $R^2$ values higher than 0.99. This suggests that the
ACTM-simulated vertical distribution of NO$_2$ has a general pattern, which can be
emulated by Gaussian functions. Once a vertical profile was constructed, it can be
easily used to estimate NO$_2$ concentration at any height.

In this study, we used the framework in Sect. 3 to estimate the OMI-derived surface
NO$_2$ concentration globally. To validate the OMI-derived surface NO$_2$ concentrations,
ground-measured surface NO$_2$ concentration in China, the US and Europe in 2014
was collected (Fig. 4). The total number of NO$_2$ observations in China, the US and
Europe are 43, 373 and 88 respectively. The OMI-derived annual average for all sites was 3.74 µg N m⁻³, which was close to the measured average (3.06 µg N m⁻³). The $R^2$ between OMI-derived surface NO₂ concentrations and ground-based NO₂ measurements was 0.75 and the RMSE was 1.23 µg N m⁻³ (Fig. 5), which is better than the modeling results by the GEOS-Chem ACTM ($R^2=0.43$, RMSE=1.93 µg N m⁻³). Satellite-based methods have the advantages of spatiotemporally continuous monitoring N₄ at a higher resolution, which helps alleviate the problem of the coarse resolution of ACTMs in estimating N₄ concentration and deposition.

**Fig. 4** Spatial distribution of measured surface NO₂ and NH₃ concentrations in 2014. For NO₂ (a), the measured data in China, the US and Europe were obtained from the NNDMN, US-EPA and EMEP, respectively; for NH₃ (b), the measured data in China, the US and Europe were obtained from the NNDMN, US-AMoN and EMEP, respectively.

**Fig. 5** Comparison between annual mean satellite-derived and ground-measured surface NO₂ concentrations (a), and comparison between annual mean modeled (by an ACTM as GEOS-Chem) and ground-measured surface NO₂ concentrations (b). The ground-based monitoring sites are shown in Fig. 4.

For NO₃⁻ and HNO₃, previous studies firstly constructed the relationship between NO₂, NO₃⁻ and HNO₃, and found a relatively high linear relationship between NO₂, NO₃⁻, and HNO₃.
and HNO$_3$ at a monthly or yearly scale. For example, Jia et al. (Jia et al., 2016) found a linear relationship between NO$_2$ and NO$_3^-$, HNO$_3$ concentration at annual scale ($R^2$=0.70). Similarly, based on the ground-based measurements in the NNDMN, a high correlation was found between surface NO$_2$ and NO$_3^-$ concentration at monthly or annual timescales (Fig. 6) (Liu et al., 2017c). Using these linear relationships and satellite-derived surface NO$_2$ concentration, the annual mean surface NO$_3^-$ and HNO$_3$ can be estimated. Alternatively, the relationship of NO$_2$, NO$_3^-$ and HNO$_3$ can also be modeled by an ACTM. For example, a strong relationship of tropospheric NO$_2$, NO$_3^-$ and HNO$_3$ column was simulated over all months by an ACTM, with the correlation ranging from 0.69 to 0.91 (Liu et al., 2017a). But, over shorter timescales, the relationship between NO$_2$, NO$_3^-$ and HNO$_3$ may be nonlinear, which we should be cautious about when estimating surface NO$_3^-$ and HNO$_3$ concentration from NO$_2$ concentration.
Fig. 6 Correlation between surface NO₂ and particulate NO₃⁻ concentration in the NNDMN at annual and monthly scales, which were adopted from Liu et al. (Liu et al., 2017c). (a) indicates the spatial locations of monitoring sites in the NNDMN; (b) and (c) represent yearly and monthly relationship between surface NO₂ and particulate NO₃⁻ concentration, respectively.

For the wet N deposition, Liu et al. (Liu et al., 2017a) followed the framework in Sect. 3 to estimate wet nitrate deposition using ABL NO₂ columns derived from OMI NO₂ column and NO₂ vertical profile from an ACTM (MOZART), and precipitation by a mixed-effects model showing the proposed model can achieve high predictive power for monthly wet nitrate deposition over China (R=0.83, RMSE=0.72).

4.2 Surface NH₃ Concentration and Reduced N Deposition

With the development of atmospheric remote sensing of NH₃, some scholars have
estimated surface NH$_3$ concentration and dry NH$_3$ deposition based on the satellite NH$_3$ column data. Assuming the ratio between the surface NH$_3$ concentration to the NH$_3$ column was fixed, Yu et al. (Yu et al., 2019) applied a linear model to convert satellite NH$_3$ columns to surface NH$_3$ concentration and estimated dry NH$_3$ deposition in China using the inferential method. But Yu et al. (Yu et al., 2019) did not consider the spatial variability of the vertical profiles of NH$_3$, which may cause a large uncertainty in estimating surface NH$_3$ concentration.

In Western Europe, Graaf et al. (Graaf et al., 2018) used the ratio of the surface NH$_3$ concentration (in the bottom layer) to total NH$_3$ column from an ACTM to convert the IASI NH$_3$ column to surface NH$_3$ concentration, and then estimated dry NH$_3$ deposition combining the modeled deposition velocity and IASI-derived surface NH$_3$ concentration. Similarly, in North America, Kharol et al. (Kharol et al., 2018) estimated the dry NH$_3$ deposition by the CrIS-derived surface NH$_3$ concentration and deposition velocity of NH$_3$. They found a relatively high correlation (R=0.76) between the CrIS-derived surface NH$_3$ concentration and AMoN measurements during warm seasons (from April to September) in 2013 (Fig. 7). Over China, Liu et al. (Liu et al., 2017b) found a higher correlation (R=0.81) between IASI-derived surface NH$_3$ concentrations and the measured surface NH$_3$ concentrations than those from an ACTM (R=0.57, Fig. 8).
Fig. 7 Comparisons of the measured surface NH$_3$ concentration by the AMoN and CrIS-derived surface NH$_3$ concentration in the US during warm season (April-September) in 2013 (Kharol et al., 2018). (a) and (b) indicate measured and CrIS-derived surface NH$_3$ concentration at the AMoN sites, respectively; (c) represents the comparison of averaged surface NH$_3$ concentration during warm months between CrIS-derived estimates and measurements, while (d) indicates the comparison of monthly surface NH$_3$ concentration between CrIS-derived estimates and measurements.

Fig. 8 Comparisons of the measured surface NH$_3$ concentration with IASI-derived surface NH$_3$ concentration at the NNDMN sites over China (Liu et al., 2017b). (a) indicates the comparison of measured and modeled surface NH$_3$ concentration from an ACTM (MOZART), and (b) represents the comparison of the measured and IASI-derived surface NH$_3$ concentration.

Liu et al. (Liu et al., 2019) followed the framework in Sect. 3 to estimate the IASI-derived surface NH$_3$ concentration (at the middle height of the first layer by an
ACTM) (Fig. 9), and found a good agreement with ground-based surface NH$_3$ concentration. The correlation between the measured and satellite-derived annual mean surface NH$_3$ concentrations over all sites was 0.87 as shown in Fig. 10, while the average satellite-derived and ground-measured surface NH$_3$ concentration was 2.52 and 2.51 μg N m$^{-3}$ in 2014 at the monitoring sites, respectively. The satellite-derived estimates achieved a better accuracy ($R^2$=0.76, RMSE = 1.50 μg N m$^{-3}$) than an ACTM (GEOS-Chem, $R^2$=0.54, RMSE = 2.14 μg N m$^{-3}$).

Surface NH$_3$ concentrations

Fig. 9 Spatially satellite-based surface NH$_3$ estimates in 2014 (Liu et al., 2019). The global surface NH$_3$ concentration datasets have been released on the website: https://zenodo.org/record/3546517#.Xj6l4GgzY2w.

Fig. 10 Comparison between yearly satellite-based and measured surface NH$_3$ concentrations (a), and comparison between yearly modeling (by an ACTM as GEOS-Chem) and measured surface NH$_3$ concentrations (b) (Liu et al., 2019). The ground-based monitoring sites are shown in Fig. 4.

The proposed methods (Liu et al., 2019) can also estimate NH$_3$ concentration at any...
height using the constructed vertical profile function of NH₃. The Gaussian function can well emulate the vertical distribution of NH₃ from an ACTM outputs with 99% of the grids having R² values higher than 0.90 (Fig. 11). This means, for regional and global estimation, the vertical distribution of NH₃ concentration has a general pattern, which can be mostly emulated by the Gaussian function. Once a global NH₃ vertical profile was simulated, it can be easily used to estimate satellite-derived NH₃ concentration at any height. We can also estimate dry NH₃ deposition using the IASI-derived surface NH₃ concentration combining the modeled Vₐ. To date, there are still no studies developing satellite-based methods to estimate the wet reduced Nᵣ deposition on a regional scale.

Fig. 11 Spatial distributions of R² for Gaussian function by simulating NH₃ and NO₂ vertical profiles. This is an example of Gaussian fitting using 47 layers’ NH₃ and NO₂ concentration from an ACTM (GEOS-Chem).

5. Trends of Surface Nᵣ Concentration and Deposition by Satellite-based Methods

The Nᵣ concentration and deposition modeled by ACTMs are highly dependent on the accuracy of input Nᵣ emissions. The methods commonly used to estimate anthropogenic Nᵣ emissions are based on the data of human activities and emission factors, which can be highly uncertain. The ACTM methods driven by Nᵣ emission inventory have relatively poor timeliness, and have limitations in monitoring the recent trends of Nᵣ deposition. Satellite-based methods provide a simple, fast and relatively objective way to monitoring Nᵣ deposition at a high resolution, and less susceptible to the errors in the
assumptions that emission inventories are compiled based on, particularly the lack of reliable data over developing countries (Crippa et al., 2018). With such advantages, researchers developed the satellite-based methods to estimate surface \( \text{Nr} \) concentration, deposition and even emissions. Satellite-based methods have advantages in monitoring the recent trends of \( \text{Nr} \) deposition. Geddes et al. (Geddes and Martin, 2017) used NO\(_2\) column from the GOME, SCIAMACHY, and GOME-2 to estimate satellite-derived NO\(_x\) emissions, and then used the calibrated NO\(_x\) emission inventory to drive an ACTM to simulate the long-term oxidized \( \text{Nr} \) deposition globally. They found oxidized \( \text{Nr} \) deposition from 1996 to 2014 decreased by 60% in Eastern US, doubled in East China, and declined by 20% in Western Europe (Fig. 12). We use the datasets by Geddes et al. (Geddes and Martin, 2017) to calculate the trends of total oxidized \( \text{Nr} \) deposition during 1996-2014. It is obvious that two completely opposite trends exist: (1) in East China with a steep increase of higher than 0.5 kg N ha\(^{-1}\) y\(^{-1}\) and (2) East US with a steep decrease of lower than -0.5 kg N ha\(^{-1}\) y\(^{-1}\). Although it is not a direct way to use satellite \( \text{Nr} \) observation to estimate \( \text{Nr} \) deposition, the method of estimating trends of \( \text{Nr} \) deposition by Geddes et al. (Geddes and Martin, 2017) can be considered effective since it took account of the changes of both NO\(_x\) emission and climate by an ACTM.

**Fig. 12** Gridded annual changes of total oxidized \( \text{Nr} \) deposition simulated by GEOS-Chem constrained with GOME, SCIAMACHY, and GOME-2 NO\(_2\) retrievals during 1996-2014 (Geddes and Martin, 2017). We gained the generated datasets.
Some researchers developed a more direct way to infer the trends of surface \( N_r \) concentration and deposition. Geddes et al. (Geddes et al., 2016) presented a comprehensive long-term global surface \( \text{NO}_2 \) concentration estimate (at 0.1° resolution using an oversampling approach) between 1996 and 2012 by using \( \text{NO}_2 \) column from the GOME, SCIAMACHY, and GOME-2. The surface \( \text{NO}_2 \) concentration in North America (the US and Canada) decreased steeply, followed by Western Europe, Japan and South Korea, while approximately tripled in China and North Korea (Geddes et al., 2016). Jia et al. (Jia et al., 2016) established a simple linear regression model based on OMI \( \text{NO}_2 \) column and ground-based surface \( N_r \) concentration, and then estimated the trends of dry \( N_r \) deposition globally between 2005 and 2014. They found that dry \( N_r \) deposition in Eastern China increased rapidly, while in the Eastern US, Western Europe, and Japan dry \( N_r \) deposition has decreased in recent decades.

We split the time span of 2005-2016 into two periods: 2005-2011 and 2011-2016, as surface \( \text{NO}_2 \) concentration shows opposite trend in China in these two periods. The magnitudes of both growth and decline in surface \( \text{NO}_2 \) concentration in China are most pronounced worldwide in the two periods (Fig. 13). During 2005-2011, apart from Eastern China with the largest increase in surface \( \text{NO}_2 \) concentration, there are also several areas with increasing trends such as Northwest and East India (New Delhi and Orissa), Western Russia, Eastern Europe (Northern Italy), Western US (Colorado and Utah), Northwestern US (Seattle and Portland), Southwestern Canada (Vancouver, Edmonton, Calgary), Northeast Pakistan and Northwest Xinjiang (Urumqi). Notably, the biggest decreases in surface \( \text{NO}_2 \) concentration during 2005-2011 occurred in Eastern US and Western EU (North France, South England, and West German).
During 2011-2016, due to the strict control of NO\textsubscript{x} emissions, Eastern China had the largest decrease in surface NO\textsubscript{2} concentration than elsewhere worldwide, followed by Western Xinjiang, Western Europe and some areas in Western Russia.

**Fig. 13** Gridded annual changes in surface NO\textsubscript{2} concentrations gained by OMI retrievals during 2005-2011 (a) and during 2011-2016 (b) in this study. We have released the global surface NO\textsubscript{2} concentrations during 2005-2016 available at the website: https://zenodo.org/record/3546517#.Xj6I4GgzY2w.

Liu et al. (Liu et al., 2019) estimated surface NH\textsubscript{3} concentration globally during 2008-2016 using satellite NH\textsubscript{3} retrievals by IASI. A large increase of surface NH\textsubscript{3} concentrations was found in Eastern China, followed by Northern Xinjiang province in China during 2008-2016 (**Fig. 14**). Satellite-based methods have been proven as an effective and unique way to monitoring the trends of global N\textsubscript{r} concentration and deposition. To date, there are still few studies reporting the satellite-derived trends of reduced N\textsubscript{r} deposition on a global scale.

**Fig. 14** Gridded annual changes in surface NH\textsubscript{3} concentrations gained by IASI retrievals during 2008-2016 (Liu et al., 2019). We have released the global surface NH\textsubscript{3} concentrations during 2008-2016 at the website: https://zenodo.org/record/3546517#.Xj6I4GgzY2w.
6. Remaining Challenges for Estimating \( N_r \) Deposition Using Satellite Observation

First, the reduced \( N_r \) deposition plays an important contribution to total \( N_r \) deposition. \( \text{NH}_3 \) exhibits bi-directional air-surface exchanges. The \( \text{NH}_3 \) compensation point (Farquhar et al., 1980) is also an important and highly variable factor controlling dry \( \text{NH}_3 \) deposition (Schrader et al., 2016; Zhang et al., 2010). However, the current existing satellite-based methods did not consider this bi-directional air-surface exchange. It is important to better parameterize the \( \text{NH}_3 \) compensation point, and assess the effects of bi-directional air-surface exchanges on estimating the dry \( \text{NH}_3 \) deposition.

Second, the existing satellite-based methods to estimate \( N_r \) deposition used the ratio of the surface \( N_r \) concentration to the \( N_r \) column by an ACTM to convert satellite \( N_r \) column to surface \( N_r \) concentration. However, the calculated ratio (by an ACTM) and the satellite \( N_r \) column have different spatial resolutions, and previous studies usually applied the modeled ratio directly or interpolate the ratio into the resolution of satellite \( N_r \) column. This method assumes the relationship at coarse resolution by an ACTM can also be effective in fine resolution as satellite indicated. When regional studies are conducted, regional ACTMs coupled with another meteorological model (e.g. WRF-Chem, WRF-CMAQ) (Grell et al., 2005; Wong et al., 2012) can be configured to match the spatial resolution of satellite observation, but this is not as viable for global ACTMs (e.g. MOZART, GEOS-Chem) due to differences in model structures and computational cost. The modeled ratio of surface \( N_r \) concentration to the \( N_r \) column may have variability at spatial scales finer than the horizontal resolution of global ACTMs. The impact of such scale effect (at different spatial scales) on estimated surface \( N_r \) concentration should be further studied.
Third, the satellite observation can only obtain reliable NO\textsubscript{2} and NH\textsubscript{3} column presently, and there are no available high-resolution and reliable direct HNO\textsubscript{3}, NO\textsubscript{3}\textsuperscript{-}, NH\textsubscript{4}\textsuperscript{+} retrievals. For HNO\textsubscript{3}, NO\textsubscript{3}\textsuperscript{-}, NH\textsubscript{4}\textsuperscript{+} concentrations, the satellite-based methods often applied the satellite-derived NO\textsubscript{2} and NH\textsubscript{3} concentration and the relationship between N\textsubscript{r} species from an ACTM (or ground-based measurements) to estimate surface HNO\textsubscript{3}, NO\textsubscript{3}\textsuperscript{-}, NH\textsubscript{4}\textsuperscript{+} concentration. With the development of satellite technology, more and more N\textsubscript{r} species can be detected, such as HNO\textsubscript{3}. However, at present, satellite HNO\textsubscript{3} products are not mature, and the spatial resolution is low. Direct, high-resolution and reliable satellite monitoring of more N\textsubscript{r} species is critical to further developing the use of using atmospheric remote sensing to estimate N\textsubscript{r} deposition at global and regional scales.

Fourth, estimating wet N\textsubscript{r} deposition using satellite NO\textsubscript{2} and NH\textsubscript{3} column remains relatively uncommon. Further studies should focus on how to combine the high-resolution satellite NO\textsubscript{2} and NH\textsubscript{3} column and the ground-based monitoring data to build wet N\textsubscript{r} deposition models to estimate wet N\textsubscript{r} deposition at higher spatiotemporal resolution. The proposed scheme to estimate the wet N\textsubscript{r} deposition in Sect. 3 is statistical. On the other hand, the wet N\textsubscript{r} deposition includes the scavenging processes of in-cloud, under-cloud and precipitation. Processed-level knowledge and models can benefit the estimation of wet N\textsubscript{r} deposition using satellite NO\textsubscript{2} and NH\textsubscript{3} column.

7. Conclusion

The recent advances of satellite-based methods for estimating surface N\textsubscript{r} concentration and deposition have been reviewed. Previous studies have focused on using satellite NO\textsubscript{2} column to estimate surface NO\textsubscript{2} concentrations and dry NO\textsubscript{2} deposition both regionally and globally. The research on calculating surface NH\textsubscript{3}
concentration and reduced N\textsubscript{r} deposition by satellite NH\textsubscript{3} data is just beginning, and some scholars have carried out estimating surface NH\textsubscript{3} concentration and dry NH\textsubscript{3} deposition on different spatial and temporal scales, but the research degree is still relatively low. We present a framework of using satellite NO\textsubscript{2} and NH\textsubscript{3} column to estimate N\textsubscript{r} deposition based on recent advances. The proposed framework of using Gaussian function to model vertical NO\textsubscript{2} and NH\textsubscript{3} profiles can be used to convert the satellite NO\textsubscript{2} and NH\textsubscript{3} column to surface NO\textsubscript{2} and NH\textsubscript{3} concentration at any height simply and quickly. The proposed framework of using satellite NO\textsubscript{2} and NH\textsubscript{3} column to estimate wet N\textsubscript{r} deposition is a statistical way, and further studies should be done from a mechanism perspective. Finally, we summarized current challenges of using satellite NO\textsubscript{2} and NH\textsubscript{3} column to estimate surface N\textsubscript{r} concentration and deposition including a lack of considering NH\textsubscript{3} bidirectional air-surface exchanges and the problem of different spatial scales between an ACTM and satellite observation.

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Data availability. OMI NO\textsubscript{2} datasets are available at http://www.temis.nl/airpollution/no2.html. IASI NH\textsubscript{3} datasets are available at https://cds-espri.ipsl.upmc.fr/etherTypo/index.php?id=1700&L=1. Surface NO\textsubscript{2} concentration during 2005-2007 obtained by Nowlan et al. (Nowlan et al., 2014) and longterm estimates (1996-2012) by Geddes et al. (Geddes et al., 2016) are available at
http://fizz.phys.dal.ca/~atmos/martin/?page_id=232. Total oxidized N₉ deposition simulated by GEOS-Chem constrained with GOME, SCIAMACHY, and GOME-2 NO₂ retrievals during 1996-2014 (Geddes and Martin, 2017) is available at http://fizz.phys.dal.ca/~atmos/martin/?page_id=1520. A database of atmospheric N₉ concentration and deposition from the nationwide monitoring network in China is available at https://www.nature.com/articles/s41597-019-0061-2. Measured N₉ concentration and deposition datasets in the United States are available on the website: https://www.epa.gov/outdoor-air-quality-data. Measured surface NO₂ and NH₃ concentration datasets in Europe are available at https://www.nilu.no/projects/ccc/emepdata.html. Global surface NO₂ and NH₃ concentration data used to calculate the longterm trends in Fig. 13 and Fig. 14 have been released on the website: https://zenodo.org/record/3546517#.Xj6I4GgzY2w.

Competing interests. The authors declare no competing financial interests.

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