



This discussion paper is/has been under review for the journal Atmospheric Measurement Techniques (AMT). Please refer to the corresponding final paper in AMT if available.

Towards validation of ammonia (NH₃) measurements from the IASI satellite

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Received: 22 September 2014 – Accepted: 11 November 2014 – Published: 4 December 2014

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Limited availability of ammonia (NH_3) observations is currently a barrier for effective monitoring of the nitrogen cycle. It prevents a full understanding of the atmospheric processes in which this trace gas is involved and therefore impedes determining its related budgets. Since the end of 2007, the Infrared Atmospheric Sounding Interferometer (IASI) satellite has been observing NH_3 from space at a high spatiotemporal resolution. This valuable data set, already used by models, still needs validation. We present here a first attempt to validate IASI- NH_3 measurements using existing independent ground-based and airborne data sets. The yearly distributions reveal similar patterns between ground-based and space-borne observations and highlight the scarcity of local NH_3 measurements as well as their spatial heterogeneity and lack of representativity. By comparison with monthly resolved data sets in Europe, China and Africa, we show that IASI- NH_3 observations are in fair agreement but that they are characterized by a smaller variation in concentrations. The use of hourly and airborne data sets to compare with IASI individual observations allows to investigate the impact of averaging as well as the representativity of independent observations for the satellite footprint. The importance of considering the latter and the added value of densely located airborne measurements at various altitudes to validate IASI- NH_3 columns are discussed. Perspectives and guidelines for future validation work on NH_3 satellite observations are presented.

1 Introduction

Ammonia (NH_3) is a key component of our ecosystems and the primary form of reactive nitrogen (Nr) in the environment (Erisman et al., 2007; Sutton et al., 2013a). It represents more than half of Nr atmospheric emissions (Galloway, 2003). Mainly released by food production, the drastic increase of NH_3 emission in the atmosphere in the last century was due to the need to feed an ever growing population combined with

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greatly increased rates of livestock production (Erisman et al., 2008; Galloway et al., 2008). This increase has numerous environmental impacts on the ecosystems (Sutton et al., 2008; Erisman et al., 2013). As the major basic species in the atmosphere, NH₃ reacts rapidly with acid gases and drives the acidity of precipitations and particulate matter (Behera et al., 2013). Its role in aerosol formation (Hertel et al., 2012) makes it an important component in air quality and climate issues (Pope III et al., 2009; Erisman et al., 2011). In addition to being directly toxic to plants at high concentrations (Krupa, 2003), NH₃ and its derivatives are also quickly deposited in the ecosystems, increasing their eutrophication and reducing biodiversity (Erisman et al., 2007; EEA-European Environment Agency, 2014). All these impacts are magnified as the nitrogen atom included in NH₃ enters the “Nitrogen Cascade” (Galloway et al., 2003).

Human activities have caused a large increase of NH₃: manure management and agricultural soils are responsible for more than 82 % of the 49.3 Tg of NH₃ emitted globally in 2008 (EDGAR-Emission Database for Global Atmospheric Research, 2011). The second largest contribution is from biomass burning, mainly linked to large scale fires (6 Tg in 2008, following EDGAR v4.2 inventory), but also to a lesser extent to agricultural waste burning (0.76 Tg in 2008). It is worth noting that global emission inventories have an uncertainty of at least 30 % (Sutton et al., 2013a). The amount of NH₃ emitted in the atmosphere is also strongly dependent on agricultural practices and climatic conditions (Sutton et al., 2013a, b) and this causes large variability in emissions on national/regional scales (Reis et al., 2009). At a local scale, other sources such as traffic and/or industry can be important (Gong et al., 2011). A further increase of NH₃ emissions is expected during this century due to agricultural intensification and the projected increases in surface temperature, which favors NH₃ volatilization (Fiore et al., 2012; Sutton et al., 2013a).

Despite the importance of NH₃ for the environment, the atmospheric processes in which it is involved and the related budgets are still poorly understood (Fowler et al., 2013). Progress in instrumentation, flux measurements and understanding of processes during the last decades have allowed advances in local/regional modeling

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(Flecharde et al., 2013). For example, the development of a bi-directional parameterization of surface/atmosphere exchange of NH₃ has improved regional modeling, for which the information needed as input (e.g. emission inventories, meteorological data) are well known (Wichink Kruit et al., 2012; Bash et al., 2013). More limitations exist on NH₃ modeling at global scale. As an example, in the multi-model comparison of Den-
tener et al. (2006), only seven on 23 models included NH_x (Fowler et al., 2013). Even if the NH₃ cycle is more and more integrated in the simulations (e.g. see Hauglustaine et al., 2014), one of the key limitations for global modelling is still the availability of measurements across the globe.

Measuring NH₃ is indeed challenging because of (i) strong temporal and spatial variability of ambient levels, (ii) quick conversion of NH₃ from one phase to another (gas/particulate/liquid) and (iii) its stickiness to the observational instruments (Sutton et al., 2008; von Bobruzki et al., 2010). There are currently very few monitoring stations that provide daily or hourly resolved NH₃ measurements, with most long-term monitoring of NH₃ being made using a passive sampler or dedicated denuder with a time resolution of several weeks. Some countries have their own network providing long term ground-based observations (e.g. United-States (US), Netherlands, United-Kingdom (UK)) but these are the exception and most of the data sets are restricted to a certain period (e.g. NitroEurope data set, Flecharde et al., 2011). The spatial coverage of these networks and campaigns is therefore strongly heterogeneous, with the large majority of available measurements in the Northern Hemisphere and an underrepresentation of other regions such as tropical agroecosystems (Bouwman et al., 2002). Airborne data sets are beginning to become available (e.g. Nowak et al., 2010; Leen et al., 2013) and provide information about the vertical distribution of NH₃. It is worth noting that a few ship campaigns (e.g. Norman and Leck, 2005; Sharma et al., 2012) have also supplied NH₃ observations in oceanic atmosphere and that on-road measurements have recently been performed at landscape scale (Sun et al., 2014). The measurement gap identified above is of special importance considering the large variability of NH₃ in time and space.

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Over the last few years satellite instruments able to detect NH₃ have begun to fill this observational gap, allowing new insights on global emissions, distributions and transport (Clarisse et al., 2009; Shephard et al., 2011; Van Damme et al., 2014a). Moreover, the spatial footprint of the satellite sounders currently available offers area-averaged measurements that are in much better correspondence with the grid cell size of current atmospheric chemistry and transport models, in comparison with the point monitoring of atmospheric concentrations made at the ground (Flechard et al., 2013). First comparisons of model results with satellite measurements at global (Clarisse et al., 2009; Shephard et al., 2011) and continental (Heald et al., 2012; Van Damme et al., 2014b) scales have been achieved and suggest an underestimation of the modeled concentrations. Satellite sounders with a high spatiotemporal resolution, such as the Infrared Atmospheric Sounding Interferometer (IASI), also offer the opportunity to identify area-specific and time-dependent emission profiles (Van Damme et al., 2014b). This would improve models that currently use a generalized and simplistic representation of the timing of emissions (van Pul et al., 2009). Satellite data are also being used in inversion methods (e.g. Zhu et al., 2013) to evaluate emission inventories at various scales (Streets et al., 2013).

While NH₃ satellite measurements have started to be used by models, their validation has yet to be performed. We present here the first attempt to validate IASI-NH₃ measurements with correlative data from the ground- and airplane-based measurements. We discuss these results critically (Sect. 3) considering the important mistime and misdistance errors (Wendt et al., 2013), which are introduced by comparing measurements of a very reactive species that are not perfectly collocated in time and space. In the next section we detail the IASI retrieval scheme implemented to derive NH₃ concentrations as well as the ground-based and airborne measurements used in this study.

2 Measurement data sets

2.1 Satellite observations

NH₃ was first detected in IASI spectra inside fire plumes above Greece in 2007 (Coheur et al., 2009). Subsequently, the development of a simplified retrieval method allowed the first global map of NH₃ from IASI observations to be produced (Clarisse et al., 2009). In Clarisse et al. (2010), a sensitivity study was performed, showing the abilities of infrared sensors to probe the lower troposphere, depending on atmospheric parameters such as the thermal contrast (see below). Recently, an improved IASI-NH₃ data set has been generated, combining better sensitivity and error characterization. The algorithm used to retrieve NH₃ columns from the radiance spectra is described in detail, as well as compared with previous ones, by Van Damme et al. (2014a). In short, the improved retrieval scheme exploits the hyperspectral characteristic of IASI and a broad spectral range between 800 and 1200 cm⁻¹. The algorithm consists of two steps. The first one is the calculation of the Hyperspectral Range Index (HRI), a dimensionless spectral index, from IASI Level1C radiance. This HRI is converted in a second step to a total column of NH₃ using look-up-tables (LUTs) built from forward radiative transfer model simulations under various atmospheric conditions. As the thermal contrast (temperature difference between the Earth's surface and the atmosphere at 1.5 km) is the critical parameter for infrared remote sensing in the lower troposphere, it is explicitly accounted for in the LUTs. Its value is derived from the temperature profile and surface temperature from the IASI Level2 information provided by the operational IASI processor (August et al., 2012). The retrieval processing also includes an error on the IASI-NH₃ total columns, taking into account the sensitivity of the satellite measurements to NH₃. This error estimate is especially important for comparisons with independent data sets (Van Damme et al., 2014b).

A known limitation of the retrieval method is that it uses only two fixed NH₃ profiles for the forward simulations (Fig. 3 in Van Damme et al., 2014a). The GEOS-Chem model was used to derive a source profile above land and a transported profile over oceans.

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The bias introduced by the use of these fixed profile shapes to build the LUTs for the total column retrieval is expected to be no higher than a factor of two on the total column values in the large majority of cases (Van Damme et al., 2014a). As independent total column data sets are not available and as the IASI-NH₃ measurements do not provide vertical information, these two modeled profiles are used in this paper to convert the retrieved column to a concentration at the surface and/or at an altitude of interest for the comparison with correlative data.

The IASI instrument is itself on-board the polar sun-synchronous MetOp platform, which crosses the equator at a mean local solar time of 9:30 a.m. and p.m. It therefore allows global retrievals of NH₃ twice a day (Clerbaux et al., 2009). In this study, we only consider the measurements from the morning overpass as they are generally more sensitive to NH₃ because of higher thermal contrast at this time of day (Van Damme et al., 2014a). Given the high spatial variability of NH₃ concentrations, the footprint of the satellite measurement is an important parameter to take into account in comparing the retrieved columns with local ground-based measurements. IASI has an elliptical footprint of 12 km by 12 km (at nadir), and up to 20 km by 39 km (off nadir) depending on the satellite viewing angle. The availability of measurements is mainly driven by the cloud coverage as only the observations with a cloud coverage lower than 25 % are processed. Van Damme et al. (2014b) have shown, given the strong dependence on thermal contrast, that spring-summer months are better suited to accurately measure NH₃ from IASI (error below 50 %). The detection limit of NH₃ depends on both thermal contrast and the vertical distribution of NH₃, an illustration of this can be found in Fig. 5 of Van Damme et al. (2014a). Note that, due to the combination of high temporal and spatial variability of this trace gas, IASI-NH₃ observations are characterized by a high coefficient of variation: 187.8 % (for measurements with a relative error below 100 %) and 85.6 % (error below 50 %) for the morning measurements above land in 2011.

2.2 Ground-based observations

Monitoring NH₃ from the ground is not straightforward due to technical limitations and to high variability of concentrations in time and space (von Bobruzki et al., 2010; Hertel et al., 2012). While the availability of NH₃ concentration and flux measurements at sub-landscape scales is increasing, measurements at landscape to regional scale are sparser (Flechard et al., 2013). In this study we use surface measurements from the US Ammonia Monitoring Network (AMoN, <http://nadp.sws.uiuc.edu/amon/>), the European Monitoring and Evaluation Programme (EMEP) network (<http://nilu.no/projects/ccc/emepdata.html>), the NitroEurope (NEU) Integrated project (<http://nitroeuropa.eu>), the Netherlands National Air Quality Monitoring Network (LML, <http://www.lml.rivm.nl>), the IGAC/DEBITS/AFRICA (IDAF) network (<http://idaf.sedoo.fr>) and a series of observations from the Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN, made available on request by X. Liu, China Agricultural University). The AMoN and EMEP data sets are used for the global comparison, the NEU and LML data sets are used for the regional assessment while NNDMN and IDAF observations are used for both. These networks and their respective characteristics are summarized in Table 1. Additional details are provided below.

The EMEP network has been running since the 1980s, with data provided by the countries as part of the Convention on Long-Range Transboundary Air Pollution. These data are made available after national and EMEP quality assessments have been conducted (EMEP, 2014). For this analysis data from 27 sites were used, which have been generated from various national networks contributing to EMEP, so that instrumentation differs from one site to another (EBAS, 2014). The AMoN network was established in 2010 in the USA to monitor NH₃ with Radiello passive diffusion-type samplers (with a time resolution of two weeks) (NADP-National Atmospheric Deposition Program, 2012). We use the 28 sites from this network which have started to provide measurements no later than 1 March 2011.

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The NEU data set was generated using DELTA (DENuder for Long-Term Atmospheric sampling) systems, detailed in Sutton et al. (2001). The network, which focused on Europe in the framework of the NEU Integrated project, is extensively described in Sutton et al. (2007) and Tang et al. (2009). Much effort was dedicated to providing a consistent data set with a high level of comparability (as the measurements were performed by different groups), including an inter-comparison study (Tang et al., 2009). It is worth noting that the precision for monthly means of a clean site using DELTA systems has been reported as below 10 % (Sutton et al., 2001). We use in our analyses the observations of 53 sites from 2008, 2009 and 2010 (Flechard et al., 2011). The Piana del Sele site was excluded as it is known to be unrepresentative for the area due to agricultural activities close by. The IDAF network is not only one of the few networks providing measurements for tropical agroecosystems, but also for the whole Southern Hemisphere. It uses passive samplers (Ferm, 1991) which have been validated in situ and are associated with a reproducibility of 14.6 % (IDAF, 2014). Measurements at the 10 IDAF sites have been considered. The data used here for 2008–2011 show a similar annual cycle with comparable amplitudes to the data from the same network covering 1998 to 2007 presented by Adon et al. (2010). The NNDMN data set for China was generated using both DELTA systems (31 sites) and ALPHA (Adapted Low-cost, Passive High Absorption) samplers (12 sites) according to the methodology of Sutton et al. (2001) and Tang et al. (2001), as detailed for this network in Liu et al. (2011). The network mainly covered farmland sites, but also included several grassland (2) and forest (4) sites across China.

Lastly, the LML network was operated using continuous annular denuder systems for the period considered here (1 January 2008–31 December 2012), based on the methodology of Wyers et al. (1993), although a switch to miniDOAS systems is planned for the near future in order to reduce running costs (Volten et al., 2012). The instrumentation used for this subset is characterized by a precision of 11 % (Wyers et al., 1993). It provides hourly NH₃ measurements at 8 sites representative for the Netherlands since 1992 (Nguyen et al., 2009).

2.3 Airborne observations

Airborne measurements offer the advantage to provide concentrations at various altitudes and typically covering a surface area that is more representative of the satellite footprint than single ground-based observations. As NH_3 emissions result primarily from ground-based sources, measurements at higher altitudes are also characterized by less spatial variability, allowing improved comparison with satellite columns. Aircraft observations are able to capture, to some extent, the intra-footprint variability spread in situ which is integrated in each satellite observation. By comparison, aircraft observations of NH_3 typically focus on short term campaign measurements.

The airborne data set used for this analysis was collected during the CalNex campaign (California Research at the Nexus of Air Quality and Climate Change), which took place in May and June 2010 in California, US (Ryerson et al., 2013; Parrish, 2014). NH_3 was measured by chemical ionization mass spectrometry (CIMS, uncertainty of $\pm(30\% + 0.2\text{ppbv})$) during 16 flights of the National Oceanic and Atmospheric Administration (NOAA) WP-3D aircraft (Nowak et al., 2007, 2010, 2012). The observations were made at 1 Hz which is equivalent to 100 m spatial resolution, with the majority of measurements at altitudes from 10 m to two kilometers above ground. Nowak et al. (2012) used this data set to show the underestimation of NH_3 emissions from dairy farms and automobiles in regional emissions inventories and to assess the impact of these sources on the formation of ammonium nitrate. Figure 1 presents the high resolution NH_3 profiles from the campaign, with x axis being linear from 0 to 100 ppbv and logarithmic from 100 to 1000 ppbv. The large majority of measurements range from 0 to 100 ppbv, with few measurements going up to several hundreds ppbv.

3 Results and discussions

This section starts with a comparison between IASI- NH_3 observations and ground-based measurements. A first qualitative NH_3 global comparison is performed for the

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year 2011, followed by quantitative regional analyses with data set over Europe, China and Africa. Then the ground-based hourly measurements provided by the Dutch national network (LML) are used to evaluate the IASI measurements collocated in time and to discuss the challenges of validating the satellite measurements of reactive species, showing high spatial and temporal variability. The added value of the airplane measurements for validation of IASI-NH₃ measurements is presented in Sect. 3.2.

3.1 Comparison with ground-based observations

3.1.1 Global evaluation

Figure 2 shows the NH₃ distribution measured by IASI for 2011 and gridded at 0.25° lat × 0.5° lon resolution for the areas covered by the available measurement data sets. In this graphic a weighting procedure has been applied on the IASI daily columns to take into account in the average the error variability of the data set (Van Damme et al., 2014a). The main agricultural source areas are observed in the Indo-Gangetic plain, the North China Plain (NCP) as well as intensive agricultural valleys such as the Ferghana Valley in Central Asia and the San Joaquin Valley in California (Clarisse et al., 2009, 2010). High NH₃ values are also seen over burned areas mainly in Eastern Europe (April) (IFFN-GFMC-16, 2011), over the Magadan region in Eastern Russia (May and especially second part of July) (NASA, 2011) and in Indonesia (March). Ground-based data sets of NH₃ concentrations for 2011 have been superimposed in Fig. 2. What is striking from the collected data sets for that year is the scarcity of the NH₃ measurements in some parts of the world. Except the IDAF stations in Africa, no measurements were available for 2011 in the rest of the tropical region and in the Southern Hemisphere. Nevertheless, we find the same general pattern in the surface measurements as in the satellite-derived distribution, with the largest concentrations for the Chinese stations, followed by the IDAF sites in Africa. Europe and USA present lower concentrations with the exception of the Eibergen station (EMEP) in the Netherlands (see also below).

while the conversion to surface observation does not change the correlation much, it determines the intercept. The use of an inadequate profile shape leads unavoidably to biased surface concentrations from IASI.

For Europe, the averaged satellite distribution for 2008, 2009 and 2010 is presented with superimposed the three year mean NEU ground-based observations (Fig. 3, top-left). High values can in particular, be seen in the western part of Russia, which is an area that was strongly impacted by the Russian fires of 2010 (R'Honi et al., 2013). The effect of these fires appears to be overrepresented in the satellite distribution due to the weighted averaging procedure associated with the higher IASI sensitivity in fire plumes (Van Damme et al., 2014a, b). Overall, however, IASI observes a similar pattern as that reported by the NEU sites. The highest surface concentrations are measured in Northwestern Europe, going up to $7 \mu\text{g m}^{-3}$ at Cabauw, Netherlands, where satellite columns are above $2 \times 10^{16} \text{ molec cm}^{-2}$. The lowest surface concentrations reported by the NEU network are observed at the Northern European stations, where IASI columns are low too. The statistical analyses (Fig. 3, bottom-left) gives a significant correlation. This is especially true considering that we are comparing monthly averaged surface concentrations from ground-based instruments (integrated over day and night) with the monthly satellite mean that includes only measurements from the morning overpass time (only monthly means with an IASI weighted relative retrieval error below 100 % have been taken into account). The linear regression is indicated in red and is characterized by a Pearson's r of 0.28 ($n = 1337$ number of coincidences), a slope of 0.42 and an intercept at $2.1 \mu\text{g m}^{-3}$. Note that if the satellite data are restricted to the monthly mean concentrations with associated error below 75 %, the Pearson's r increases to 0.35 ($n = 961$). Almost all the sites in Europe are characterized by a positive correlation coefficient when it is significant (p value below 0.05). The highest correlation ($r = 0.81$) is obtained for the Fyodorovskoye site (RU-Fyo), and this is explained by the very large columns observed during the Russian fire event of 2010. The second site characterized by a high correlation coefficient is the Monte Bondone site (IT-MBo) with $r = 0.71$, which is characterized by high NH_3 concentrations in summer and low in winter, consis-

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tent with local livestock grazing patterns. By contrast, the most northern sites, located in Finland together give an indication of negative correlation (Hyytiälä, $r = -0.44$; Lompolojänkkä, $r = -0.43$; Sodankylä, $r = -0.45$) with an associated p value slightly above 0.05. These are the sites with very low ground level concentrations (three year average close to $0.1 \mu\text{g m}^{-3}$), suggesting that the monthly variability for such small concentrations in a region associated with low thermal contrast, cannot be reliably detected by IASI. The other NEU stations with a significant r above 0.5 include Brasschaat (BE-Bra, 0.54), Fougères (FR-Fou, 0.58) and Vall d'Alinyà (ES-VDA, 0.58). The list of stations and their respective correlation coefficient, as well as their associated p and n values, are provided in Table A1.

For China (2009 to 2013), the spatial patterns in NH₃ abundance are in good agreement (Fig. 3, top-middle panel). We find the highest concentrations in the NCP area and the lowest in remote areas such as the Linzhi site (LZ, on the Tibetan Plateau) and several sites in Northeast China. To better illustrate the potential for high degree of correspondence between the satellite columns and the surface measurements we provide in Fig. 4 the monthly averaged time series at Shangzhuang (SZ) from the ground and from space ($r = 0.72$). This suburban station northwest of Beijing (40.14°N , 116.18°E), surrounded by agriculture and far from a main road (Shen et al., 2011), has one of the longest record of NH₃ concentrations in China, from January 2008 until February 2014. The IASI derived total columns (top, blue, molec cm^{-2}) reproduce nicely the ground-based concentrations (bottom, red, $\mu\text{g m}^{-3}$) time evolution with concentration peaks in summer (June–July), close to $25 \mu\text{g m}^{-3}$ at the surface and around $3 \times 10^{16} \text{ molec cm}^{-2}$ observed from IASI. Looking at inter-annual variability, for example 2013, the fertilization peaks of March, July and October are clearly observed (Shen et al., 2011). It is interesting to note that the highest January surface concentration ($5.1 \mu\text{g m}^{-3}$) and satellite total column ($7.8 \times 10^{15} \text{ molec cm}^{-2}$) are measured during the severe winter pollution episode of 2013 (see Boynard et al., 2014; Wang et al., 2014). The correlation analysis for the monthly values of the 43 NNDMN sites gives a Pearson's r equal to 0.39 (considering the IASI data with a monthly relative retrieval error below

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100 %) and a slope of the regression equal to 0.21 ($n = 1149$). The Pearson's r is globally higher than for Europe, probably due to the higher concentrations in China. This is also observed in the monthly retrieval error which are overall lower. In addition to Shangzhuang, the sites with a significant Pearson's r above 0.5 are: China Agricultural University (CAU, 0.51), Lingshandao (LSD, 0.55), Changdao (CD, 0.58), Gongzhuling (GZL, 0.52), Bayinbuluke (BYBLK, 0.59), Fengyang (FY, 0.64), Ziyang (ZY, 0.58), Yanting (YT, 0.70) and Dianchi (DC, 0.64), where the significance of r values for these sites are all less than $P = 0.01$. Only one site in the NNDMN network gives a significant negative correlation, which is the cleanest site in the network in Tibetan Plateau (LZ, $r = -0.75$). With a mean ground-based concentration of $1.03 \mu\text{g m}^{-3}$, as suggested with the Finnish sites, this again illustrates the limitation of IASI to provide reliable monthly NH₃ values at very low concentrations. The results for all the sites are presented in Table A2.

For Africa, both IASI and surface network report the highest values in West and Central Africa and the lowest in South Africa for the four year average (2008 to 2011, Fig. 3 top-right panel). However, over Western Africa the highest vmr are reported by the network for the dry savanna sites (located above 10°N) while IASI is observing higher columns closer to the coast. This could possibly be due to the lack of IASI sampling above these Sahelian sites during the rainy season in June–July, which is characterized by large concentrations measured at the surface. The high density of livestock concentrated on the fresh pasture at that time implies high surface emissions pulses associated with a high deposition and/or cloud coverage, preventing IASI to capture these events, while they are monitored from the ground (Adon et al., 2010). Adon et al. (2010) have shown that the dry savanna sites are characterized by higher concentrations during the wet season while (June–July), conversely, the wet savanna sites (closer to the West-African coast) present the larger amount during the dry season (January–February). The latter may therefore be better monitored by IASI. The right-bottom plot (Fig. 3) compares the surface vmr for Africa in ppbv. There is no correlation when the data of all the sites are grouped. It could be explained by the sparsity of

the IDAF dataset, combined with the difficulties of the satellite to catch specific high emission events. The only station showing a positive significant Pearson's r is Lamto (Ivory Coast, 0.39), which is not surprisingly a wet savanna site. The comparison for each station individually is provided in Table A3.

In general, the regression intercept is relatively high, while the slope is relatively low. For all three regions the largest measured concentrations are underestimated in the IASI surface derived data. This smaller range of concentrations could be a consequence of using a fixed vertical profile shape in the retrieval procedure for IASI spectra. Indeed, in the case of a low boundary layer height, concentrations are larger in that narrow layer, which is not considered by the straightforward procedure applied to retrieve IASI surface concentrations from the columns. Moreover, the vertical profiles above land from GEOS-Chem model have already been identified as being too low above California, especially for strongly polluted areas (Schiferl et al., 2014). Another reason could lie in the influence of very local sources on the ground-based data (see further). Contrary to what we find for high concentrations, there is a positive bias of IASI for low concentrations. There are at least three possible reasons for this. The first one stems from the way the relative retrieval errors are taken into account. Applying weighted averaging of IASI columns using the relative retrieval error tends to favor the largest concentrations as explained in Van Damme et al. (2014a). A second reason, in the case of very low NH_3 amounts, is that the random error of the HRI always translates into a positive contribution of the column (and hence does not average to zero for zero NH_3). The third reason is again linked with a misrepresentation of NH_3 vertical distribution. In low NH_3 abundance cases and/or in areas where NH_3 is being deposited, there is no strong vertical gradient near the surface. The use of a polluted profile shape (peaking at the surface) in the retrieval procedure of IASI surface concentrations tends to overestimate the surface contribution of the total columns.

We recall that all regressions shown and discussed in this section are based on weighted monthly IASI-derived NH_3 means, with the weight being determined by the relative error of the individual retrieved columns. We also made the comparisons

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with unweighted monthly means and found somewhat different parameters for the regression line but an overall weak impact on the regression coefficients. In all cases the slope remains well below 0.5 and the intercept smaller but still positive.

5 Hourly resolved data set

The LML data set offers long term hourly resolved NH_3 measurements allowing to investigate individual IASI observations and testing the effect of averaging different time scales. It is worth noting however that this network provides measurements in an area (the Netherlands) that is not particularly favorable for infrared remote sensing of surface pollution (low thermal contrast and relatively high cloud coverage). Table 2 summarizes the results from the comparison of the IASI concentrations (columns converted to surface concentrations as above) to those measured from the ground. The upper part presents the comparison from the linear regression for each individual site (slope (a) and intercept (b) of the linear regression, Pearson's r (r , underlined when significant) and the number of observations (n)). We took into account only the IASI measurements covering the stations, which means that the true elliptical footprint is considered and only the observations including the LML site are kept. Six of the few sites show significant correlation and for all, except for one (Valthermond, which have negative intercept and fairly high slope), we find a very low slope (0.03–0.3) but a high intercept, between $0.89 \mu\text{g m}^{-3}$ (De Zilk) to 1.91 (Wekerom). This shows again the difficulties of IASI to capture the entire range of local surface concentrations, as explained above. The best Pearson's r are observed for Valthermond (0.49), De Zilk (0.39) and Eibergen (0.38) sites. When all the sites are taken together we obtain a Pearson's r of 0.29, a slope of 0.07 and an intercept of $1.68 \mu\text{g m}^{-3}$. These results, obtained by comparing individual IASI value with ground-based measurement at the overpass time of the satellite, could be due as discussed previously to the misrepresentation of the vertical distribution in the processing of the spectra and the need to consider the boundary layer height.

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A factor preventing one from drawing strong conclusions from this comparison is the spatial representativity of the ground-based measurement within the large footprint of the satellite (going from 113 km² at nadir to around 613 km² for the highest satellite viewing angles). It means that local sources inside the satellite footprint could have a strong influence on the IASI column values, but not be represented in the ground-based data sets due to the high horizontal gradient of NH₃ concentrations around the source (Fowler et al., 1998; Sutton et al., 1998; Hertel et al., 2012). Conversely, background concentrations within the IASI pixel will tend to mask single local sources. The representation issue of ground-based measurement stations has already been highlighted during model performance evaluation (Wichink Kruit et al., 2012). To avoid this uncertainty which is difficult to assess in the validation, we could consider the general rule that the LML network is representative for the entire Netherlands (Nguyen et al., 2009). For instance, using the eight stations together, we can calculate a daily mean concentration of NH₃ at the overpass time of the satellite that we compare with the IASI weighted mean of the measurements inside the area covered by the network (considered here as the following box: 51.185–53.022° N and 4.01–7.106° E). Doing so, we find that the Pearson's *r* increases to 0.28 while the slope and the intercept of the linear regression are respectively 0.21 and 0.96 μg m⁻³.

Finally, to investigate the impact of temporal variability and to compare with the previous results of monthly resolved data, we calculated monthly average of IASI measurements above the Netherlands and the LML ground-based concentrations at the IASI overpass time. The time series of Fig. 5 show the temporal variability in 2008, 2009, 2010, 2011, and 2012 of IASI-NH₃ columns (top panel, blue, molec cm⁻²) and of the surface concentrations derived from IASI columns (bottom panel, red, μg m⁻³) and monitored by the LML sites (bottom panel, black, μg m⁻³). Only values with a relative retrieval error below 100 % have been taken into account. The spring peak linked with fertilization practices in the Netherlands is clearly identifiable in the LML and IASI time series in March/April. Looking at the surface concentrations, a fair agreement is found between the ground-based and the satellite observations. The monthly variability is

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consistent between both instruments, even if the IASI-derived values are not reproduc-
 ing with the same amplitude the peaks observed from the ground. This could be due, as
 explained previously, to a misrepresentation of the NH₃ vertical distribution. As shown
 in the lowest row of Table 2, the comparison based on monthly data is characterized
 by a Pearson's r of 0.47, substantially higher than the ones obtained with the previous
 data sets. The linear regression has a slope of 0.19 and an intercept of 1.86 $\mu\text{g m}^{-3}$.

3.2 Comparison with airborne observations

California is an important NH₃ source area in US. Major sources include livestock oper-
 ations, agricultural fertilizers, waste management facilities and motor vehicles (Parrish,
 2014). Figure 6 presents the NH₃ distribution for the area and period covered by the
 CalNex campaign (from 30 April to 24 June 2010). The airborne distribution (left panel,
 ppbv) is highly consistent with the satellite one (right panel, $\times 10^{16}$ molec cm⁻²). The
 retrieval error distribution is also presented as inset (bottom-left of right panel, %). The
 pattern observed is characterized by the highest concentrations in the San Joaquin
 and Sacramento Valleys, as well as in the South Coast Air Basin (SCAB). High con-
 centrations are also measured in the Imperial Valley and to a lesser extent in the neigh-
 borhood of Phoenix. Note that the high values reported by the airborne measurements
 on 22 June above Western upper part of Colorado are also measured by IASI.

To compare satellite observations with the CalNex data set, the IASI total columns
 are first converted to concentrations at the altitude of interest for each airborne mea-
 surement located inside the satellite footprint. The concentrations (airborne or satellite
 based) covering the footprint of IASI are then averaged in a second step. This results in
 222 corresponding couples of observations, on which we can apply thresholds on the
 mistime, the retrieval error or the number of airborne observations by IASI footprint.

As an illustration, Fig. 7 shows observations in the SCAB (satellite footprint centered
 at 34.12° N and 118° W) characterized by a satellite mean concentration of 2.17 ppbv
 (associated with a retrieval relative error of 25 %) and an airborne concentration of
 2.65 ppbv. This subset of observations has been selected as the one with the highest

density of airborne measurements for a given IASI footprint (with a relative retrieval error below 100 %) considering a mistime below three hours. The IASI footprint in that case includes 1515 individual airborne observations spanning altitudes from 272 m to 2.6 km.

The results obtained from the comparison are presented in Table 3. The first row gives the statistical parameters for all the coupled observations. This comparison is characterized by a Pearson's r of 0.36 ($n = 222$), a slope of 0.07 and an intercept at 2.33 ppbv. Only considering couples of observations with a mistime of less than three hours, the Pearson's r increases significantly to 0.72 ($n = 75$). If a threshold on the retrieval relative errors is also taken into account (in this case being $< 100\%$), the r increases further to 0.82. Finally, if we constrain the mistime to be lower than one hour, the Pearson's r reaches 0.99 ($n = 15$). With increased filtering, a decrease of the intercept is also observed. However, it is notable that the slope of the regression does not change substantially, remaining at around 0.17 to 0.18, indicating underestimates NH_3 vmr compared with the aircraft measurements using CIMS.

4 Conclusions and perspectives for the validation

Overall, IASI satellite measurements of NH_3 are consistent with the available data sets used in this study. This paper presents only the first steps to validate IASI- NH_3 columns. As shown, it is not straightforward and depends strongly on data availability, as well as on their representativity for satellite observations. The yearly distributions reveal consistent patterns and highlight the scarcity and the spatial heterogeneity of NH_3 observations from the ground, with very few measurements in the Southern Hemisphere and tropical agroecosystems. Comparisons with monthly resolved data sets in Europe, China and Africa have shown that IASI- NH_3 derived concentrations are in fair agreement considering that the ground-based observations used here for comparison are mainly monthly integrated measurements, while the satellite monthly weighted means are only taking into account observations at the morning overpass time.

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The comparison between IASI-derived concentrations and the surface concentrations measured locally was assessed using linear regressions. Statistically significant correlations were found at several sites, but low slopes and high intercepts were calculated in all cases. Overall, this points to a too small range in the IASI surface concentrations, which could partly be due to the lack of representativity of the point surface measurements in the large IASI pixel. Another possible reason for the difference lies in the use of a simple profile shape in the retrieval procedure, which does not take into account variations in the boundary layer height and hence in the mixing of pollution close to the surface. As shown in previous studies, accounting for realistic boundary layer heights in the retrieval of tropospheric column from satellite measurements improves on the comparison with surface measurements (e.g. see Boersma et al., 2009). A third possible contributing reason for the large intercept and low slope is a bias in the IASI values derived from weighted averaging, where more weight is given to data with the least uncertainty. Under clean conditions, when there is a weak signal, this tends to give more weight to high values, which partly explain the high intercept values.

In addition to the comparison with ground-based measurements, this paper has also provided a comparison of the IASI derived vmr to vertically resolved CIMS measurements from the NOAA WP-3D airplane during the CalNex campaign in 2010. The main advantage of such a comparison lies in the fact that numerous airborne measurements are available within the satellite footprint, strongly reducing spatial representativity issues. Moreover, airborne measurements are performed at altitudes that are more inline with the sensitivity of infrared nadir sounders. Consequently, a much higher correlation between IASI and the airplanes is found, e.g. characterized by a correlation coefficient of 0.82 for subset of observations associated with a retrieval error below 100 % for IASI and a mistime below three hours. Low slopes and large intercept values from the linear regressions are nevertheless obtained, similar as with the ground-based measurements, further suggesting a poor representativity of the profile shape used for IASI retrievals for these high pollution regions.

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More generally, this first attempt to validate the NH₃ measurements from IASI has highlighted known limitations in both the retrieval procedure (fixed vertical profile and boundary layer height) and in the correlative measurements, which are in many cases not representative of what the satellite measures horizontally and vertically. All statistical results were shown for concentrations (or vmr) and do not allow the validity of the IASI derived columns to be assessed, which is – considering the limited vertical sensitivity achievable – the most relevant quantity available from the satellite measurements. Therefore here we highlight the need to acquire more comprehensive data sets of NH₃ columns, e.g. by using boundary layer heights and NH₃ profile measurements (and/or estimates) to infer the columns abundance from the surface observations. For this purpose, dedicated measurement campaigns focusing on the NH₃ vertical profile, or additional total column measurements from ground-based Fourier-Transform InfraRed (FTIR), which are starting to be available (Vigouroux et al., 2013), will no doubt allow improvements in the validation of NH₃ satellite retrieved columns in the future.

Acknowledgements. IASI has been developed and built under the responsibility of the “Centre National d’Etudes Spatiales” (CNES, France). It is flown on-board the Metop satellites as part of the EUMETSAT Polar System. The IASI L1 data are received through the EUMETCast near real-time data distribution service. The research in Belgium was funded by the F.R.S.-FNRS, the Belgian State Federal Office for Scientific, Technical and Cultural Affairs (Prodex arrangement 4000111403 IASI.FLOW). M. Van Damme is grateful to the “Fonds pour la Formation à la Recherche dans l’Industrie et dans l’Agriculture” of Belgium for a Ph.D. grant (Boursier FRIA). L. Clarisse and P.-F. Coheur are respectively Research Associate (Chercheur Qualifié) and Senior Research Associate (Maître de Recherches) with F.R.S.-FNRS. C. Clerbaux is grateful to CNES for scientific collaboration and financial support. We gratefully acknowledge support from the project “Effects of Climate Change on Air Pollution Impacts and Response Strategies for European Ecosystems” (ÉCLAIRE), funded under the EC 7th Framework Programme (Grant Agreement No. 282910). Part of this research was supported by the EC under the 7th Framework Programme, for the project “Partnership with China on Space Data (PANDA)”. We also would like to thanks S. Bauduin, J. Hadji-Lazaro and J.-L. Lacour as well as R. van Oss, H. Volten and D. Swart for their helpful advices.

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Table 2. Statistical analysis of the LML and IASI satellite surface concentrations comparison in the Netherlands from 1 January 2008 until 31 December 2012. The spatial averaging (two bottom rows in the table) consists in considering the mean of all the ground-based observations and the one of the satellite observations in the area covered by the LML network (considered here as the box 51.185–53.022° N and 4.01–7.106° E). The temporal averaging (bottom row) consists in taking the mean of the observations (at the overpass time of the satellite) on a monthly basis. The slope and the intercept of the linear regression are indicated by a and b while the significant Pearson's correlation coefficients (r) are underlined (p value below 0.05); n corresponds to the number of points. Only IASI derived values with a relative retrieval error below 100 % have been taken into account.

Site (lat-lon)	a	b	r	n
Vredepeel (51.541° N–5.854° E)	0.04	2.49	0.16	94
Huijbergen (51.435° N–4.360° E)	0.3	1.08	<u>0.31</u>	54
De Zilk (52.298° N–4.510° E)	0.16	0.89	<u>0.39</u>	45
Wieringerwerf (52.805° N–5.051° E)	0.03	1.26	<u>0.11</u>	91
Zegveld (52.139° N–4.838° E)	0.07	1.38	<u>0.21</u>	93
Eibergen (52.096° N–6.606° E)	0.12	1.46	<u>0.38</u>	102
Wekerom (52.113° N–5.709° E)	0.03	1.91	<u>0.25</u>	87
Valthermond (52.877° N–6.931° E)	0.63	–1.39	<u>0.49</u>	78
all sites individually	0.07	1.68	<u>0.29</u>	644
all sites spatially averaged	0.21	0.96	<u>0.28</u>	960
all sites spatially and temporally averaged	0.19	1.86	<u>0.47</u>	58

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Table A1. Continued.

Site code	Site name	lat (° N)	lon (° E)	<i>r</i>	<i>p</i> value	<i>n</i>
DE-Meh	Mehrstedt	51.28	10.66	0.26	0.20	25
PT-Mi1	Mitra II (Evora)	38.54	-8.00	-0.11	0.58	29
IT-MBo	Monte Boone	46.03	11.08	0.71	0.00	36
SE-Nor	Norua	60.08	17.47	-0.19	0.37	25
CH-Oe1	Oensingen	47.29	7.73	0.03	0.89	30
UA-Pet	Petrodolinskoye	46.50	30.30	0.35	0.05	30
PL-wet	Polwet	52.76	16.31	-0.21	0.28	28
FR-Pue	Puechabon	43.74	3.60	0.09	0.63	35
IT-Ren	Renon	46.59	11.43	-0.05	0.79	33
DK-Lva	Rimi	55.70	12.12	0.54	0.11	10
DK-Ris	Risbyholm	55.53	12.10	0.54	0.27	6
IT-Ro2	Roccarespampani	42.39	11.92	0.06	0.72	33
IT-SRo	San Rossore	43.73	10.28	0.03	0.85	32
SE-Sk2	Skyttorp	60.13	17.84	-0.40	0.25	10
FI-Sod	Sodankylä	67.36	26.64	-0.41	0.07	20
DK-Sor	Soroe	55.49	11.65	-0.05	0.82	24
NL-Spe	Speulderbos	52.25	5.69	0.10	0.57	33
DE-Tha	Tharat	50.96	13.57	0.44	0.02	30
ES-VDA	Vall d'Alinyà	42.15	1.45	0.58	0.00	35
BE-Vie	Vielsalm	50.31	6.00	0.11	0.65	21
DE-Wet	Wetzstein	50.45	11.46	0.47	0.01	28

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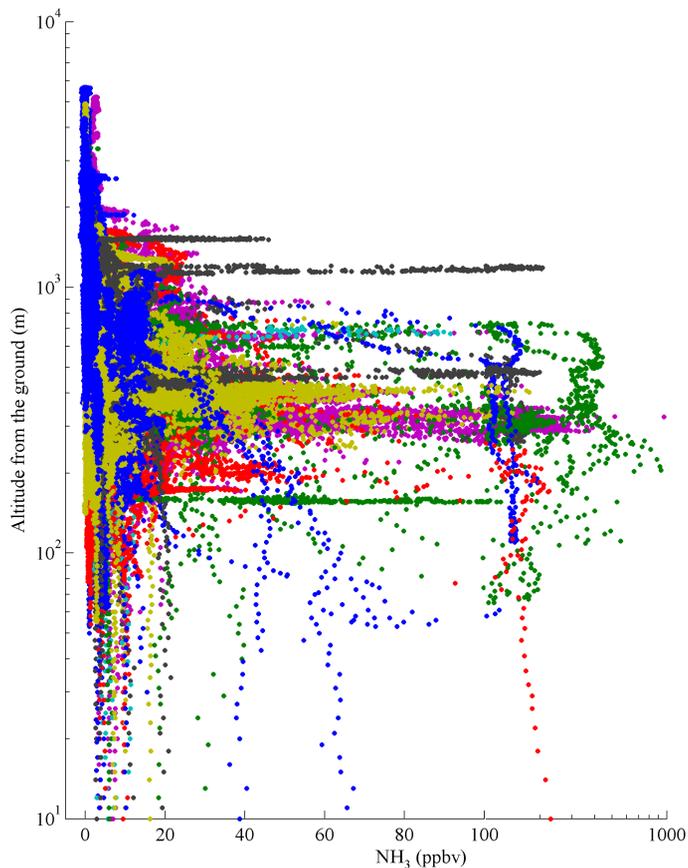


Figure 1. Vertical distribution of NH₃ airborne measurements from the NOAA WP-3D flights during the CalNex campaign in California (2010). Each color corresponds to one day of measurement.

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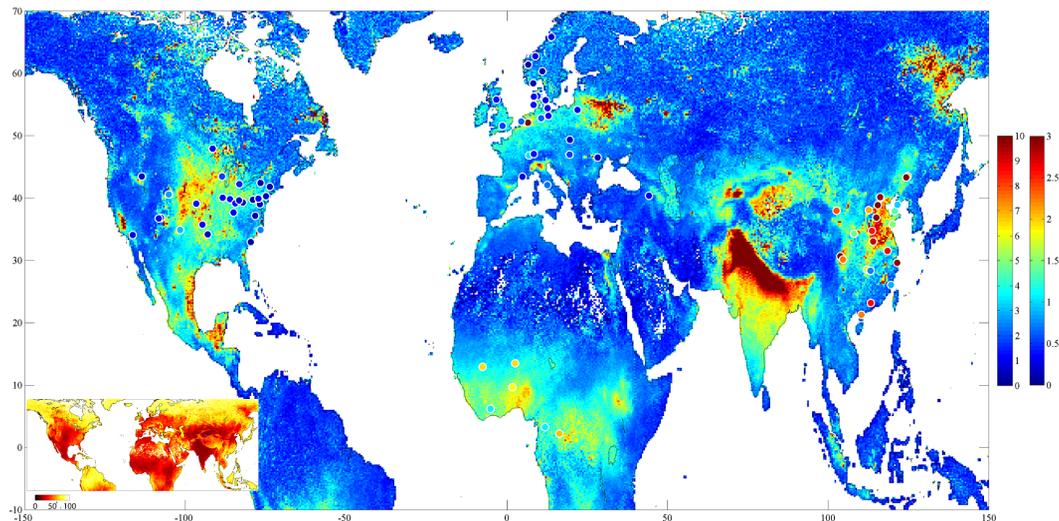


Figure 2. Yearly averaged surface concentrations ($\mu\text{g m}^{-3}$, left vertical color bar) from IDAF, AMoN, EMEP and NNDMN data sets plotted on top of the NH₃ IASI satellite columns ($\times 10^{16}$ molec cm⁻², right vertical color bar) distribution for 2011 gridded at 0.25° lat \times 0.5° lon. Columns and relative error (% , bottom-left inset) have been calculated as a weighted mean of all IASI measurements within a cell, following equations described in Van Damme et al. (2014a) (columns with an associated relative above 100 % have been filtered).

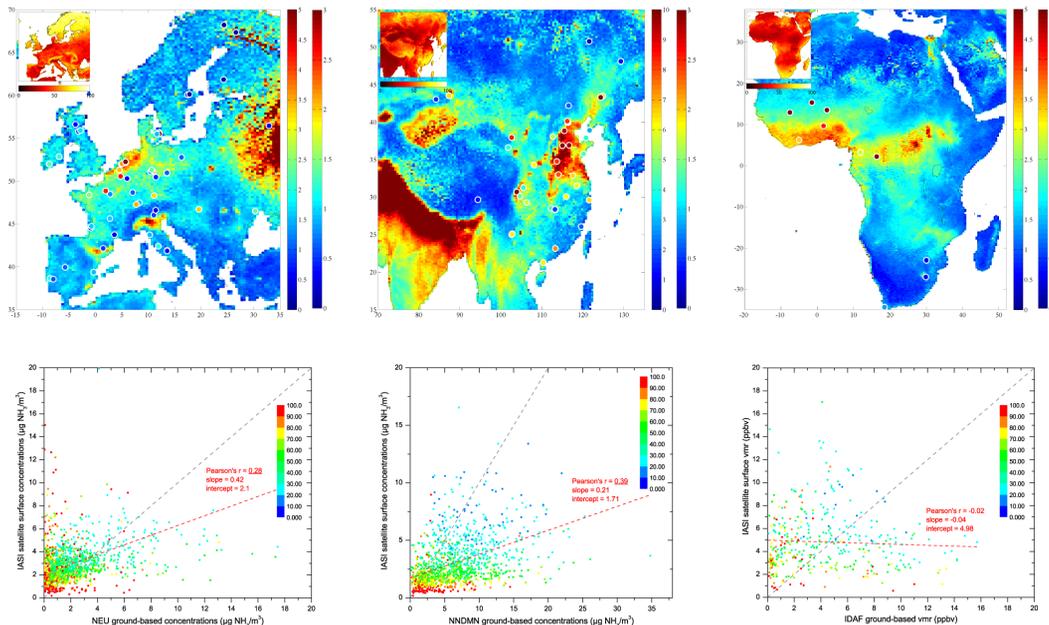
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Figure 3. Top: ground-based quantities (left vertical color bar) from NEU ($\mu\text{g m}^{-3}$, left panel), NNDMN ($\mu\text{g m}^{-3}$, middle panel) and IDAF (ppbv, right panel) data sets plotted on top of the NH₃ satellite columns ($\times 10^{16}$ molec cm⁻², right vertical color bar) distribution gridded at 0.25° lat \times 0.5° lon, both averaged for the period covered by the datasets. Stations with less than two third of the monthly concentrations available have been excluded. The relative error distribution from IASI retrieval is shown as inset (top-left, %, horizontal color bar). Columns/errors have been calculated as a mean of all the measurements within a cell, weighted by the relative error following equations described in Van Damme et al. (2014a). Bottom: monthly NEU (left panel), NNDMN (middle panel) ground-based concentrations ($\mu\text{g m}^{-3}$) and IDAF (right panel) ground-based vmr (ppbv) vs. IASI retrieved NH₃ surface concentrations ($\mu\text{g m}^{-3}$) or vmr (ppbv). The color scale corresponds to the IASI retrieval errors (%). Dashed red lines represent the linear regression and gray dashed lines the one to one line. The Pearson's r (underlined when significant (p value below 0.05)), slope and intercept are also indicated in red.

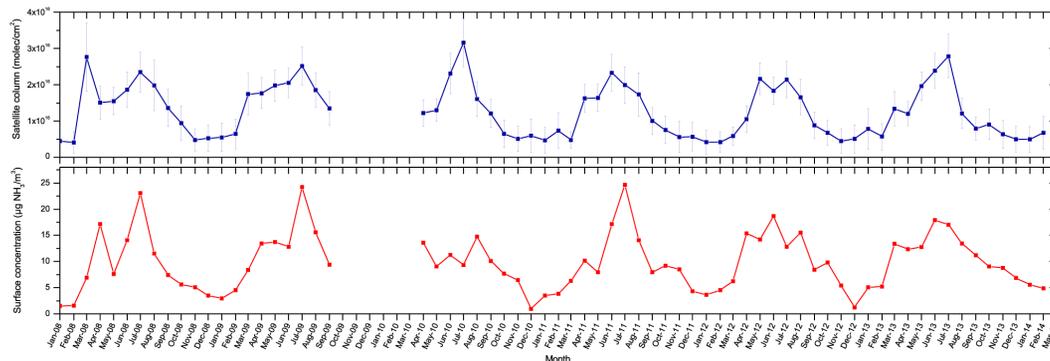


Figure 4. Time series of monthly averaged NH₃ satellite columns (top, blue, molec cm⁻²) and ground-based concentrations (bottom, red, µg m⁻³) at Shangzhuang site (40.14° N, 116.18° E) in the North China Plain (NCP) from January 2008 until February 2014. The associated weighted mean error for each month is calculated following equations described in Van Damme et al. (2014a) and presented here as error bars.

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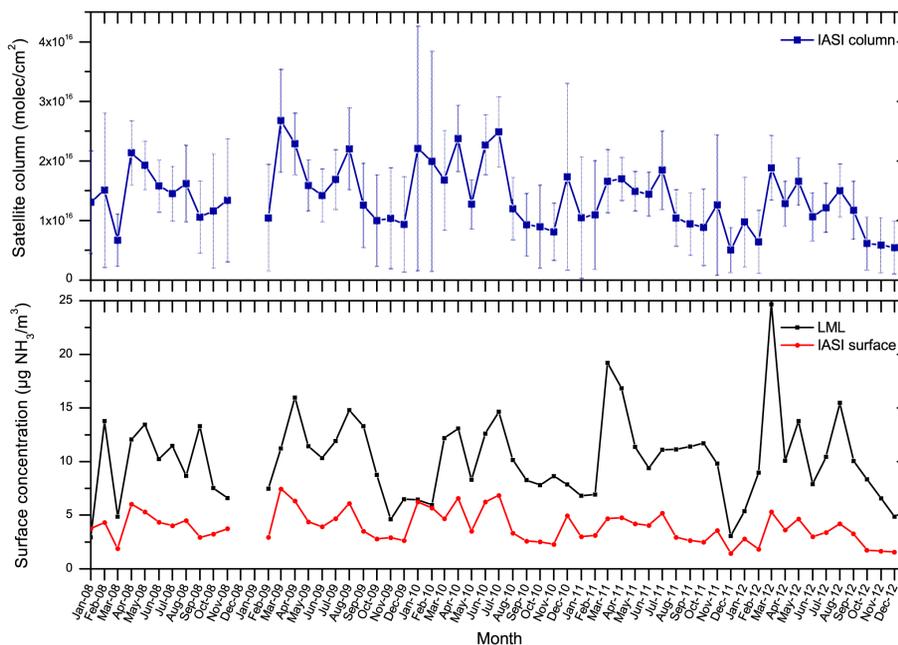



Figure 5. Monthly time series of NH₃ satellite columns (top, blue, molec cm⁻²) and surface concentrations at the overpass time of the satellite (bottom) from all the sites of the Dutch LML network averaged (black, µg m⁻³) and from IASI satellite observations (red, µg m⁻³) (in the box 51.185–53.022° N and 4.01–7.106° E, corresponding to the area covered by the Dutch network), covering 2008, 2009, 2010, 2011 and 2012. The associated error bars are based on the retrieval error for each monthly column calculated following equations described in Van Damme et al. (2014a). Only monthly means associated with weighted relative retrieval errors below 100 % are shown.

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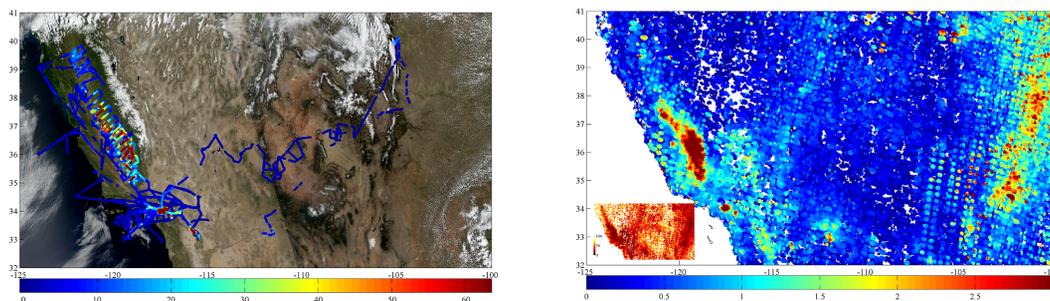



Figure 6. NH₃ distribution measured from CIMS instrument on-board NOAA WP-3D aircraft (left, ppbv) and IASI satellite (right, molec cm⁻²) averaged for the flight period of the Calnex campaign (from 30 April 2010 to 24 June 2010). Averaged relative retrieval error (%) of the IASI-NH₃ columns are presented as inset in the lower left corner of right panel. Only columns mean associated with weighted relative retrieval errors below 100% are shown.

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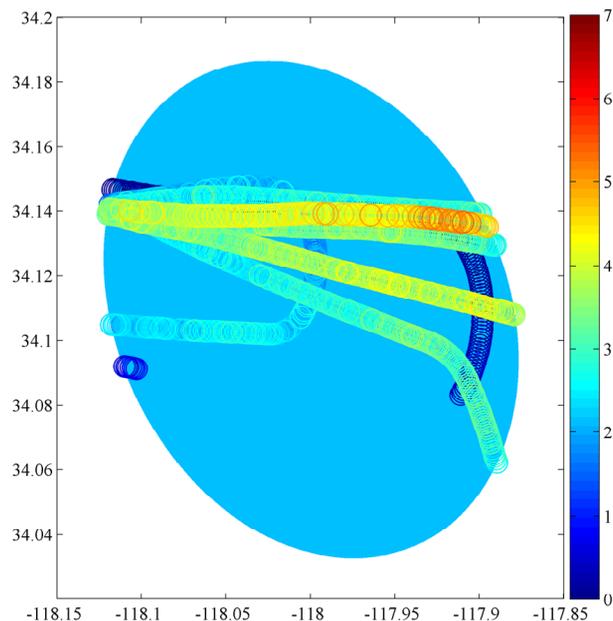


Figure 7. Spatial distribution of 1515 airborne observations made on-board the NOAA WP-3D aircraft at various altitudes superimposed on the corresponding IASI footprint. The color scale corresponds to the individual concentrations (ppbv) measured by the CIMS instrument while the value for IASI is the average of the vmr calculated from the same total column at the different altitudes of the aircraft.

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