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Temporal and spatial variability of atmospheric ammonia in the Lombardy region (Northern Italy)

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Abstract. This work investigates the spatial and temporal variability of atmospheric ammonia concentrations in the Lombardy region in Northern Italy, where its continuous measurement at hourly resolution began in 2007 at monitoring sites representative of three different land use areas (urban, rural, and mountain areas). Ammonia concentration data have been jointly elaborated with wind direction and speed to highlight the association between the origin of the air masses and the concentration levels observed at the monitoring sites far from the primary sources, essentially consisting of farming activities and cattle and pigs breeding activities located in the South-Eastern part of the region. The annual average concentrations of ammonia observed at urban (4-13 µg m^{-3} range) and rural (17-35 µg m^{-3} range) monitoring sites are in substantial agreement with literature data, which are however limited and strongly influenced by the measurement techniques used. The lowest concentration levels (0.4-5 µg m^{-3} range) are observed at the monitoring sites in the mountain areas. Both the seasonal and daily time patterns of the concentrations appear strongly related to the features of the measurement sites, namely with regard to the monitoring sites most exposed to emissions of agricultural activities, whose seasonal practices determine emissions responsible for strong variations in the ammonia atmospheric presence. Conversely, in the mountain areas in the North of region, weather conditions of atmospheric circulation seem to play a more important role than local sources, with the highest concentrations occurring when the breezes transport ammonia-rich air masses from the Southern part of the region.

Keywords: Ammonia, Time pattern, Polar plots, Po valley

1. Introduction

The interest for atmospheric ammonia (NH_3) is linked to its fundamental role in the processes of ecosystems acidification and of water eutrophication originated by its deposition on sensitive environments (Zhang et al., 2012; Bobbink et al., 1998). Furthermore, NH_3 is the main basic compound capable of neutralizing atmospheric acid gases and thus is a precursor of secondary inorganic particulate material, whose effects are well known harmful to the environment and human health (Schlesinger and Cassee, 2003; Erisman and Schaap, 2004; Sutton et al., 2009). NH_3 can react with sulphur dioxide (SO_2) and nitrogen oxides (NO_x) to form ammonium sulphate ($(NH_4)_2SO_4$), ammonium bisulphate (NH_4HSO_4), and ammonium nitrate (NH_4NO_3), significantly contributing to fine particle mass (PM2.5) with relevant implications for human health (Brunekreef and Holgate, 2002), for atmospheric visibility and for global radiation budgets (Clarisse et al., 2009; Horvath, 1992; Sutton et al., 1994). Modeling simulations (Heald et al., 2012; Schiferl et al., 2014) and air quality monitoring data (Gong et al., 2013) confirmed the specific role of NH_3 in the formation of secondary inorganic aerosol, with negative effects on both climate and air quality (Erisman et al., 2013; Paulot and Jacob, 2014). In spite of its environmental significance, atmospheric NH_3 still receives scarce attention and is not subject to air

quality standards. In the European Union atmospheric emission limits are fixed only for very few sources, such as waste incinerators where NH_3 is used in both catalytic and non-catalytic processes for NO_x emission control. However, national NH_3 emission reduction commitments for Member States are set by the National Emission Ceilings Directive for 2020 and 2030 (EU Directive 2016/2284/EU).

At the global scale NH_3 emissions have more than doubled since pre-industrial times because of both agricultural intensification and widespread use of fertilizer (Galloway et al., 2003). Actually, agricultural and livestock activities represent the largest and almost exclusive NH_3 source (Warner et al., 2016); people and traffic emissions may have an impact only in the non-agricultural regions, as suggested by the higher NH_3 ambient levels found in high densely populated areas (Suh et al., 1995). In the United States more than 82% of NH_3 emissions are attributable to the agricultural sector (US EPA National Emission Inventory, 2014) with a growing trend due to the combined effect of the increase of farming and animal husbandry and the use of nitrogen fertilizers. In the European Union, the agricultural sector is responsible for more than 94% of NH_3 emissions (EEA, 2017), but the limitations on the use of synthetic nitrogen fertilizers and the improvement of practices in the management of livestock waste have instead led to a slight emission reduction. However, increasing ambient concentration trends for NH_3 have been estimated in the orders of 2.6% yr^{-1} over the US, of 1.8% yr^{-1} over the European Union, and 2.3% yr^{-1} over China (Warner et al., 2017) also in consequence of the progressive decrease of acid gases in the atmosphere, in particular SO_2, whose neutralization is an important pathway for the removal of ammonia.

Because of the combination of unfavourable morphology and climatology of the area and of the high density of emission sources, the river Po valley in Northern Italy is one of the European areas with critical issues for the values of the air quality indexes in general and also for the concentration levels of ammonia, making it one of the main NH_3 hot-spots worldwide (Figure 1, left panel). In particular, since also in the Lombardy region NH_3 emissions derive almost exclusively from agriculture, the major concern for atmospheric NH_3 is in the southernmost portion of the region, due to the high emission levels determined by the local agricultural and animal breeding activities (Figure 1, right panel).

Focusing on the Lombardy region, where for ten years the air quality monitoring network has been continuously collecting ambient NH_3 data, this work analyses the spatial variability and temporal evolution of its concentration levels, as well as the relationship between the observed concentrations and the features of the local anemological regime (wind speed and direction) in order to investigate the location of the sources that determine the most relevant impacts on air quality.

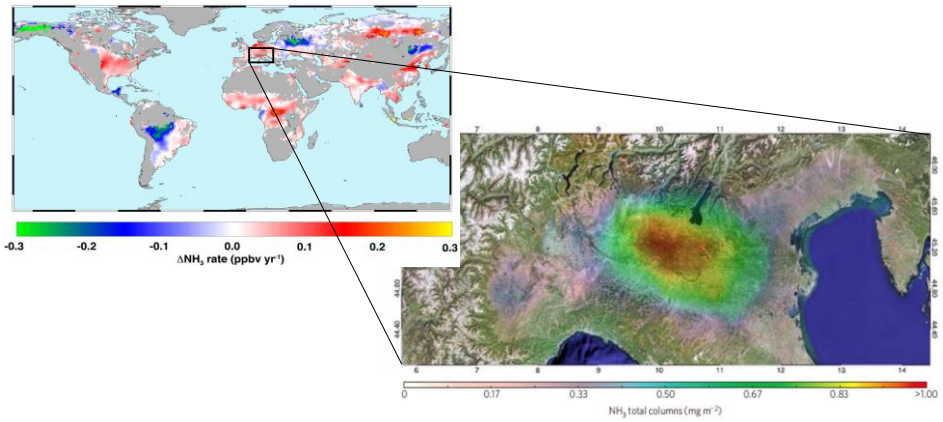

Figure 1. Left: annual change of NH_3 emission in 2002-2016 estimated from AIRS (Atmospheric Infrared Sounder, NASA) satellite measurements (Warner et al., 2017); right: average NH_3 concentration over Northern Italy in 2008 from IASI (Infrared Atmospheric Sounding Interferometer, MetOp satellite) satellite measurements (Clarisse et al., 2009)

2. Material and methods

2.1. NH_3 monitoring network in Lombardy

The air quality monitoring network operated by the Environmental Agency (ARPA) of the Lombardy region has begun to continuously collect data for atmospheric NH_3 at four sites in 2007. Progressively, the monitoring network has expanded and atmospheric NH_3 is currently measured at 11 stations, distributed almost all region at representative sites for air quality assessment, with generally very good annual data availability (more than 80% on annual basis, usually).

NH_3 concentrations are measured at hourly resolution with instruments based on the chemiluminescence technique that enables to determine NH_3 values indirectly. The measurement technique involves a sequential process with the catalytic transformation to NO of all the nitrogen compounds (NO, NO_2, and NH_3) present in the air sample and the subsequent measurement of the total nitrogen N_t (expressed as NO). Thanks to the concurrent separate measurements of both NO and NO_2 (still expressed as NO), NH_3 concentration is obtained by difference, even taking into account the efficiency of the NH_3 catalytic conversion to NO. Such analytical determination is however complex and the final NH_3 ammonia may be inaccurate just because of the critical issues of the chemiluminescence technique (Villena et al., 2012; Dunlea et al., 2007) as related to the catalytic conversion efficiencies of the various compounds, in particular for NH_3 oxidation to NO (Capiaghi et al., 2014).

2.2. NH_3 datasets

In this work NH_3 data from 12 monitoring sites collected between 2007 and 2016 have been analyzed (Figure 2). Sites have been grouped together in three areas (urban, rural and mountain), according to the classification criteria adopted by ARPA for air quality monitoring sites. In particular, the urban group (sites S1-S5) includes the two sites in the city of Cremona (360000 inhab.), those in the metropolitan area of Milan (3200000 inhab.), and the site in the city of Pavia (70000 inhab.). In Cremona the monitoring sites are located North-Eastern of the city centre (S1-CR Fatebenefratelli), and on the outskirts of the city (S2-CR Gerre Borghi), close to the banks of the Po and potentially more exposed to the emissions generated in the surrounding countryside. In the metropolitan area of Milan sites are located in the heart of the city park of Monza (S3-Monza Parco), therefore not directly exposed to emission sources, and at an urban background site in the university area of Milan (S4-MI Pascal). In Pavia the monitoring site is located at the entrance of the city park North of the city centre (S5-Pavia-Folperti). The monitoring sites of the rural group (S6-S10) are all located in the Southern part of the region. In particular, S6-Corte de' Cortesi and S8-Bertonico site are placed in a territorial context which exposes them to the emissions of agricultural and livestock activities. Actually, S6 site is in a fairly isolated location, far from roads and industrial activities, but immediately near to a pig farm; S7 and S8 sites are in areas devoted to agricultural (cereals and fodder production) and livestock activities (beef and dairy cattle and pigs). The mountain group includes the sites of Colico (S11) and Moggio (S12). Actually, S11-Colico site is located in the last town on the Eastern shore of the Lake of Como, at about 250 m a.s.l.; thanks to its location the site is not exposed to ammonia sources but may be somewhat affected by transport phenomena of pollutants emitted in the plain area of the region because of the cyclic circulation of air masses that go up the Lake Como daily. Conversely, S12-Moggio site is really in a mountain area at about 1200 m a.s.l. in Valsassina valley, a pre-alpine area where there are no significant emission sources of ammonia. Nevertheless, as for S11 site, air masses coming from the plain, located to the South, can be channeled in the valley and reach the site affecting the local air quality. Wind speed and wind direction data used for bivariate analyses in order to investigate the relationship between the observed NH_3 concentrations of the characteristics of the local anemological regime, when not concurrently available at the air quality monitoring sites, have been taken from the nearest meteorological stations, as shown in Figure 2.

Figure 2. NH_3 monitoring sites (red circles) and meteorological stations for wind data (blue squares)

3. Results

3.1. NH_3 concentration levels

The range values for the annual average concentrations at the NH_3 monitoring sites of Lombardy are summarized in Table 1. At the sites of the urban group the minimum and maximum values are in the 4-8 µg m^{-3} range and 10-13.5 µg m^{-3} range, respectively. The urban sites, mostly located in urban background environments and not directly exposed to primary emissions, show significantly lower concentrations than those observed at the rural sites, and in particular of the sites (S6-Corte de' Cortesi, S7-Piadena, and S8-Bertonico) located in areas with a strong agricultural and animal husbandry vocation, where the annual average concentrations vary from a minimum of 21.9 µg m^{-3} to a maximum of 81.7 µg m^{-3}. Interestingly, at the other two rural sites (S9 and S10) NH_3 levels are more similar to those of the urban sites, varying between a minimum of 4.8 µg m^{-3} to a maximum of 15.5 µg m^{-3}.

The annual values observed at S6 site (41.5-81.7 µg m^{-3}), practically twice as high as those of sites S7 and S8 (17.7-35.8 µg m^{-3}), highlight the impact on air quality of the pig farm located at less than 100 m from the site itself: Thus, data from this site cannot be considered representative of an area with a generic rural vocation but of the local impact of NH_3 emissions from a very specific kind of human activity. On the other hand, the lower values observed at the S9 and S10 sites suggest that, despite their location in rural areas of the Lombardy plain, they are located in areas with a less intensive agricultural vocation and far from direct emission sources. The two sites of the mountain group display the lowest annual average concentrations, between 0.4 µg m^{-3} and 5.8 µg m^{-3}, with maximum values substantially similar to the minimum values of urban sites. Thanks to their geographical position, especially for the S12-site, situated at relatively high altitude, in an area not densely populated and far from local emissive sources, the measured values can therefore be considered representative of the background level of ammonia in the atmosphere.

The time patterns of the annual average concentrations over the study period show irregular trends at the rural sites: from 2012 a strongly decreasing values are observed at S6 (from 70-80 μg m^{-3} down to 40-45 μg m^{-3}), conversely a constant increase is observed at S8 (from 20 μg m^{-3} up to 30 μg m^{-3}). At the urban sites and at the other rural sites the trends remain irregular and sometimes opposite, but with much smaller fluctuations; in general, the annual average concentrations at urban sites are around 10 μg m^{-3} with a substantially stable temporal trend. A decreasing trend is observed instead for the mountain site S12-Moggio, with the annual average concentrations in the most recent past years getting down to a few μg m^{-3}.

Table 1. Range for NH_3 annual average concentration (μg m^{-3}, at 20 °C and 101.3 kPa) in Lombardy

Area	Monitoring site	Altitude (m a.s.l.)	Dataset	NH_3 annual average
Urban	S1-CR Fatebenefratelli	43	2011-2016	4.4 - 10.7
	S2-CR Gerre Borghi	36	2012-2016	8.2 - 13.2
	S3-Monza Parco	181	2013-2016	6 - 13.4
	S4-MI Pascal	122	2007-2016	5.8 - 12.6
	S5-PV Folperti	77	2013-2016	8.1 - 13.3
Rural	S6-Corte de' Cortesi	57	2007-2016	41.5 - 81.7
	S7-Piadena	30	2013-2014	17.7 - 23.5
	S8-Bertonico	65	2009-2016	21.9 - 35.8
	S9-Schivenoglia	12	2013-2016	11.3 - 15.5
	S10-Sannazzaro	87	2013-2016	4.8 - 7.6
Mountain	S11-Colico	229	2013-2016	0.4 - 5.8
	S12-Moggio	1194	2007-2016	0.4 - 4.1

The annual average levels measured at the sites are in substantial agreement with literature data, although the comparison is purely indicative as the methods of measurement, the temporal resolution and frequency of measurements may differ also noticeably. Nevertheless, especially for the urban sites of Lombardy, the NH_3 levels values are in line with those of similar sites, such as 5.4 μg m^{-3} reported for Seoul in South Korea (Phan et al., 2013) and 15.9 μg m^{-3} for Beijing in China (Meng et al., 2011). A substantial agreement is also observed for the rural sites, except for the S6-Corte de' Cortesi site, whose peculiarity has already been discussed. As for the sites in this study, both relatively low values, in the orders of 4-6 μg m^{-3} (Wang et al., 2015; Zbieranowski and Aherne 2013; Meng et al., 2011) as at S10 site, and higher values, in the orders of 14-22 μg m^{-3} (Erisman et al., 2001, Shen et al., 2011) as at sites S7, S8, and S9, are reported in the literature for rural sites worldwide.

3.2. Bivariate analyses

The association between the NH_3 concentration levels and the local features of the anemological regime has been investigated by means of graphic representations obtained by the joint analysis of the hourly NH_3 concentration and direction and wind speed data. In the resulting polar plot representations, the angle with respect to the y axis represents the direction of origin of the wind with respect to the North, the distance from the center, the speed of the wind, the color the average value of NH_3 concentration detected under such wind conditions (Carslaw and Ropkins, 2012). In practice, the color maps allow the concentration levels observed to be associated with certain wind conditions, thus providing indications on the location and distance of the emission sources that influence the quality of the air at the monitoring site. These representations can thus account for the presence of single sources (i.e.: large industrial complexes), highlighting the situations in which they are located upwind of the monitoring site, as well as for the presence of diffuse sources homogeneously distributed around the monitoring site (i.e.:

domestic heating in urban areas), for which, on the other hand, there is no clear association between high levels of concentration and wind direction.

This last case applies to the polar plots of this study that in general do not indicate a determining role of the wind but rather confirm the widespread nature of NH_3 emissions. However, the plots highlight some peculiarities of the different measurement sites. For the rural site S6, for example, in addition to a significant seasonal variability of NH_3 levels, the highest concentration associated to weak winds confirm the role of the nearby pig farm as a relevant local emission source. Furthermore, the substantial circular symmetry of the color plots reflects the location of the site at a point surrounded by agricultural land (Figure 3). Similar considerations apply to the S8 rural site, where however the effect of the local source responsible for the maximum values observed at site S6 is missing. The plots for the urban site in Milan also show a circular symmetry in correspondence of weak winds (Figure 4), but they show, regardless of the season, the lowest NH_3 concentration values for North-North Westerly strong winds (> 4 m s^{-1}). These conditions occur during foehn episodes, actually quite rare on an annual basis (7-10 events), which bring clean air masses from the Alps down to the plain of Lombardy, lowering the concentration levels of all the atmospheric pollutants (Mira-Salama et al., 2008).

Conversely, the association of high NH_3 concentration values with medium-intensity winds blowing from East to East South East in autumn suggests transport phenomena from the Southern part of the region, where concentration levels are particularly high, towards the foothills area of the Alps. The presence of these transport phenomena is also highlighted in the summer plot of the mountain site S12, where the highest concentrations are observed concurrently with winds blowing from South West (Figure 5). As already reported for other pollutants (Dosio et al., 2002), the masses of air moving towards the North are channeled along the Eastern branch of Lake Como, and from there to the Valsassina valley, bringing with it the ammonia emitted from agricultural activities in the plain area of Lombardy. In winter, this phenomenon is not evident and apparently we observe, instead, a separation between the air masses that insist on the plain and those of the mountain area, favored also by the frequent phenomena of thermal inversion typical of the Po valley during the cold seasons.

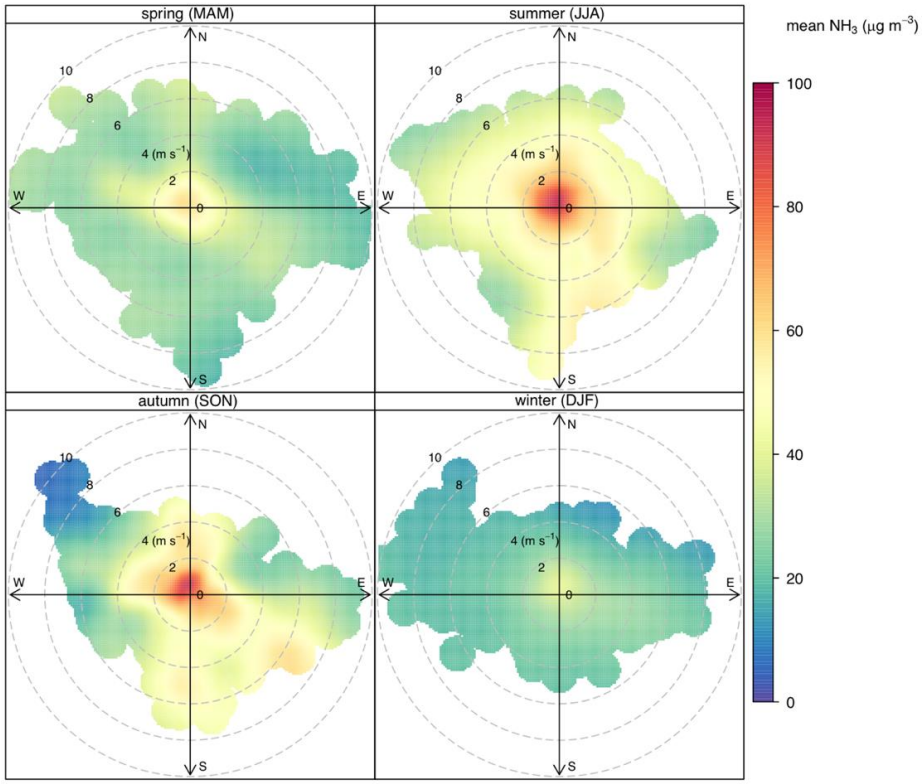

Figure 3. Seasonal polar plots for NH_3 hourly concentrations at rural site S6

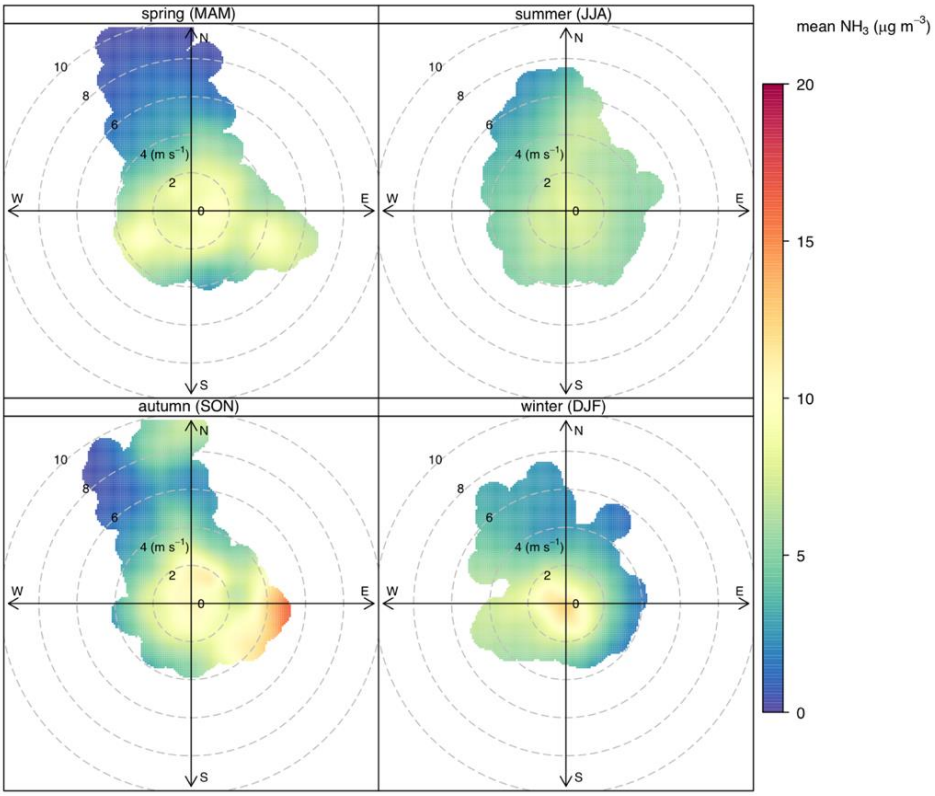

Figure 4. Seasonal polar plots for NH_3 hourly concentrations at urban site S4

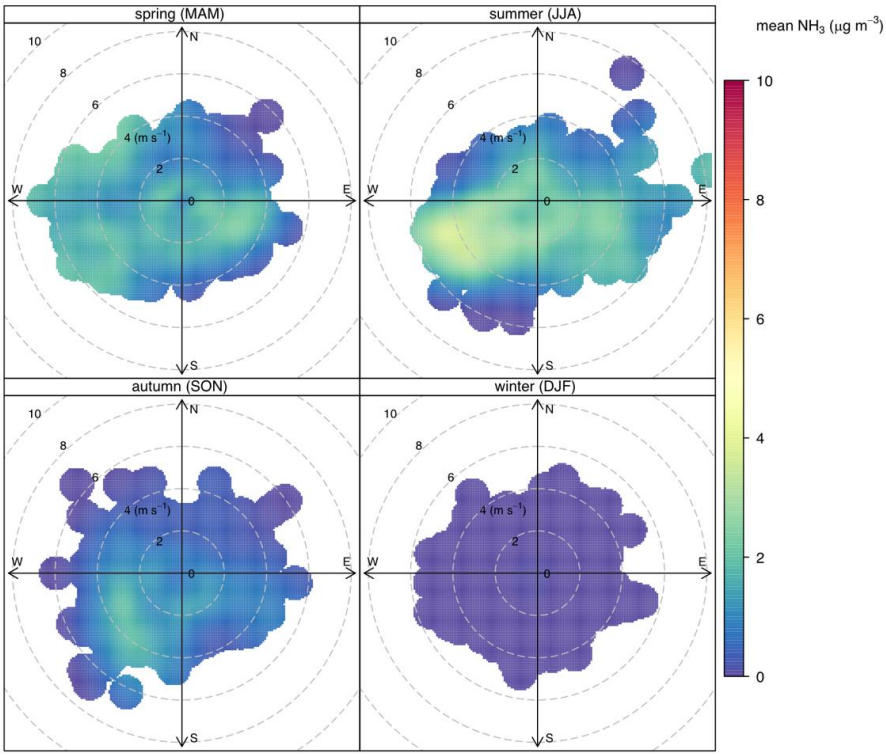

Figure 5. Seasonal polar plots for NH_3 hourly concentrations at mountain site S12

The association between high NH_3 concentrations and given wind conditions highlighted by the polar plots is further confirmed by the conditional wind roses for hourly concentration values higher than the 90th percentile of the entire NH_3 data series. This analysis confirms the presence of transport phenomena of NH_3-rich air masses from the Southern part of the region to the northernmost areas. In particular, for the two sites in Milan metropolitan area the highest concentrations are observed more frequently in correspondence of South-easterly winds, while in the case of S12 mountain site of Moggio for winds blowing from West-South West.

4. Conclusions

In the Lombardy region atmospheric NH_3 levels show a marked spatial and temporal variability with the highest concentrations in rural areas (annual averages in the orders of 20-80 µg m^{-3}) and the lowest in at mountain sites (annual average less than 6 µg m^{-3}). At urban sites, the annual average concentrations are usually around 10 µg m^{-3} with a substantially stable time trend from 2007. The NH_3 levels observed in Lombardy are substantially in agreement with literature data for similar sites, even though data are still limited and strongly influenced by the measurement techniques used.

The temporal pattern of concentrations is strongly dependent on the features of the measurement sites, namely for the rural sites more directly exposed to the emissions of agricultural activities. Seasonal practices of agricultural activities determine local emissions responsible for strong variations in atmospheric NH_3 concentration levels, especially in early spring and autumn. This variability, although attenuated, is also found at urban sites, confirming the absolutely dominant role of the emissions of the agricultural and zootechnical sector, already highlighted by the emission inventories. Furthermore, the joint analysis of the concentration data local wind conditions has highlighted the association between concentration levels and the origin of air masses, showing that the highest concentration values in urban

areas occur when the breezes carry NH_3-rich air masses from the areas at high emission intensity of the Po Valley.

5. References

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