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Swedish National Nitrogen Budget - Atmosphere

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In cooperation with SMHI

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Preface

IVL Swedish Environmental Research Institute has together with the Swedish Meteorological and Hydrological Institute (SMHI) quantified flows of reactive nitrogen to and from the atmosphere above Sweden including the atmosphere above the Swedish coastal waters. Calculations were done for year 2015. This work has been conducted with financial support from the Swedish Environmental Protection Agency as a step towards establishing a complete Swedish national budget for reactive nitrogen.

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Summary

In this report, pool 7 'Atmosphere' (AT) of the Swedish National Nitrogen Budget (NNB) is presented. The atmospheric pool of NNB is one of the 8 major pools defined by the Task Force of Reactive Nitrogen (TFRN) which together represent a total national nitrogen budget capturing all major flows of all forms of reactive nitrogen (Nr) within a country and across the country borders. The methodology to calculate NNB has been provided by the Expert Panel on Nitrogen Budgets (EPNB) in the Annexes to the ECE/EB.AIR/119 – "Guidance document on national nitrogen budgets".

The major flows of Nr to and from the tropospheric air masses above Sweden, including the Swedish national coastal waters, were calculated for the year 2015. Except for the small amount of Nr generated by lightnings, the atmospheric Nr pool is dominated by four main flows: the import and export of Nr by transboundary transport; by Swedish emissions of Nr; and by atmospheric deposition.

The principal source of the emission data has been the official Swedish reporting to the Centre on Emission Inventories and Projections (CEIP) under the Air convention (The Convention on Long-range Transboundary Air Pollution, CLRTAP). For the reactive nitrogen deposition in Sweden the official national operational estimates by the MATCH Sweden system were used.

The results show that in 2015 Sweden was a net importer of air pollution in the form of Nr, i.e. the import of Nr from the emission sources outside the country (139 kt N) was larger than the export (96.1 kt N). The Swedish emissions of Nr in 2015 were in total 117 kt N. The largest emissions come from two sectors: Agriculture (43.4 kt N) and Energy & Fuels (40.6 kt N). The deposition of Nr to Swedish ecosystems in 2015 was 160 kt N, of which 87% originated from emission sources abroad.

Sammanfattning

I denna rapport presenteras pool 7 ”Atmosfär” (AT) som ingår i Sveriges nationella kvävebudget (National Nitrogen Budget, NNB). Atmosfärspoolen i Sveriges NNB är en av de totalt 8 pooler som definierats av Task Force on Reactive Nitrogen (TFRN) som tillsammans representerar en komplett nationell kvävebudget. En fullständig NNB innefattar en kvantifiering av alla större flöden av alla former av reaktivt kväve (Nr) inom landet och över landsgränserna. Metoden för att beräkna NNB har tillhandahållits av expertpanelen för kvävebudgetar (Expert Panel on Nitrogen Budgets, EPNB) i bilagorna till ECE/EB.AIR/119 – "Guidance document on national nitrogen budgets".

De största flödena av Nr till och från de troposfäriska luftmassorna ovanför Sverige, inklusive de svenska nationella kustvattnen, beräknades för år 2015. Förutom en mindre mängd Nr som genereras av blixtar domineras den atmosfäriska Nr-poolen av fyra huvudflöden: import och export av Nr från gränsöverskridande transport; svenska utsläpp av Nr; och atmosfäriskt nedfall.

Den huvudsakliga källan till utsläppsdata har varit den officiella svenska rapporteringen till Centre on Emission Inventories and Projections (CEIP) under luftkonventionen (Convention on Long-range Transboundary Air Pollution, CLRTAP). För det reaktiva kvävedenedfallet i Sverige användes de officiella nationella beräkningarna från MATCH Sverigesystemet.

Resultaten visar att Sverige under 2015 var en nettoimportör av luftföroreningar i form av Nr, det vill säga att importen av Nr från utsläppskällorna utanför landet (139 kt N) var större än exporten (96,1 kt N). De svenska utsläppen av Nr 2015 var totalt 117 kt N. De största utsläppen kommer från två sektorer: Jordbruk (43,4 kt N) och Energi & Bränsle (40,6 kt N). Depositionen av Nr på svenska ekosystem år 2015 var 160 kt N, varav 87 procent härrörde från utsläppskällor utomlands.

Introduction

The Task Force on Reactive Nitrogen (TFRN) was established under the Working Group on Strategies and Review (WGSR) by the Executive Body at its twenty-fifth session in December 2007.

The purpose of TFRN has been defined as: *"The Task Force will develop in the long-term technical and scientific information and options which can be used for strategy development across the UNECE to encourage coordination of air pollution policies on nitrogen in the context of the nitrogen cycle and which may be used by other bodies outside the Convention in consideration of other control measures."* For the full terms of reference of the Task Force, see Executive Body decision 2007/1.

At the first meeting held in Wageningen in 2008 TFRN agreed to define reactive nitrogen (Nr) as all biologically active, photochemically reactive and radiatively active N compounds in the biosphere and atmosphere. This meant, in practice, all N except N_2 gas; for example, nitric oxide (NO), nitrogen dioxide (NO_2), nitrate (NO_3^-), organic N compounds, nitrous oxide (N_2O), ammonia (NH_3) and ammonium (NH_4^+). At the same meeting it was proposed that an expert panel could help in preparing for the reporting of national budgets, first exploring methodologies and providing a reference template for the compilation. The Expert Panel on Nitrogen Budgets (EPNB) was established (first as an ad-hoc group) and commenced work to prepare guidelines for compilations of national N budgets of individual countries. EPNB prepared the "Guidance Document on National Nitrogen Budgets". The document was presented and approved at the 31st meeting of the Executive Body of the Convention on Long-Range Transboundary Air Pollution in December 2012. The document can be downloaded from http://www.clrtap-tfrn.org/sites/clrtap-tfrn.org/files/documents/EPNB_new/ECE_EB.AIR_119_ENG.pdf. After that, the work of EPNB continued to provide detailed guidelines for each of the 8 main parts of the National N Budget (NNB) summarised in Annexes to the ECE/EB.AIR/119 – "Guidance document on national nitrogen budgets". Currently the version dated 02. 03. 2021 is available at http://www.clrtap-tfrn.org/sites/clrtap-tfrn.org/files/documents/EPNB_new/EPNB_annex_20210302_public.pdf and it contains seven out of the eight pools, the Waste annex is still under development.

There have been attempts constructing nitrogen budgets in some of the European countries and elsewhere, see for example Switzerland (Heldstab et al., 2010 and 2013), Germany (Geupel et al., 2009), Denmark (Hutchings et al., 2014) or Canada (Clair et al., 2014). These budgets have not followed the same protocol when constructed but provide information on important flows. In Europe, Sutton et al. (2011) estimated that 74% of the total input of reactive nitrogen to the environment stems from the Haber-Bosch process, 16% from combustion, and the remaining 10% from biological fixation, import of feed and products. Leip et al. (2011) estimated nitrogen fluxes for EU27 developing and using the same protocol for all countries. Leip et al. (2011) recommend development of nitrogen budgets nationwide since the assessment and management of the budgets could become an effective tool to prioritize measures and prevent unwanted effects.

National nitrogen budgets (NNB) following the EPBN methodology are constructed based on eight pools. In this report, data on pool 7 (Atmosphere, NNB_AT) are presented. The Swedish data presented in this report are for year 2015.

National nitrogen budget (NNB) for Atmosphere (pool 7)

The atmosphere in terms of N-budgets mainly functions as a transport medium, as it serves to collect, to deposit and to transport reactive nitrogen under various chemical forms in the troposphere. Main input flows are atmospheric import of Nr, and emissions from all other pools in a National Nitrogen Budget (NNB) (Cited from the EPNB_annex 7 Atmosphere). Output flows are export of Nr by atmospheric transport and Nr-deposition to land-based pools. N_2 fixation is not an outflow from the atmosphere pool, since N_2 is not considered reactive.

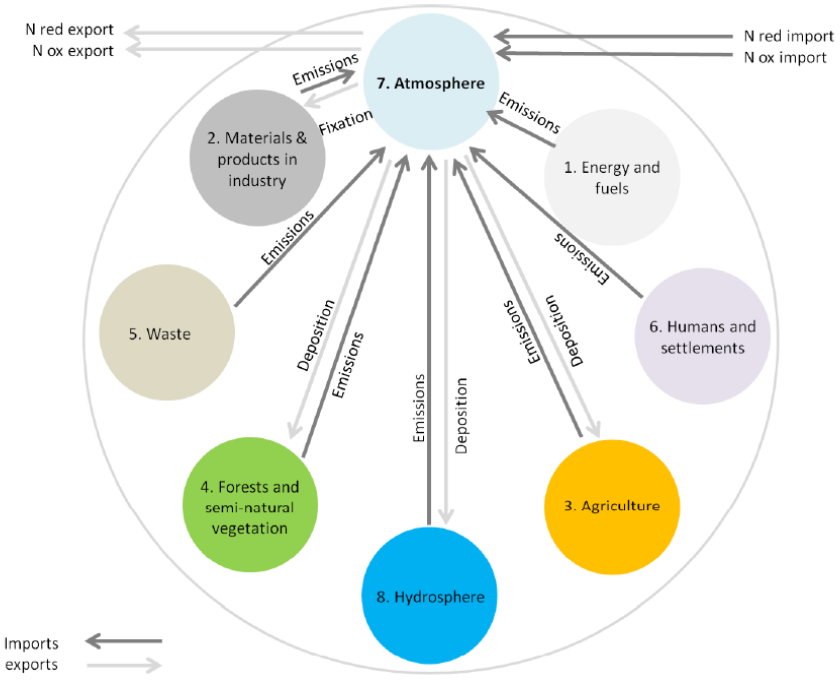

Figure 1. N budget scheme for the pool 'Atmosphere'. Source: http://www.clrtap-tfrn.org/sites/clrtap-tfrn.org/files/documents/EPNB_new/EPNB_annex_20210302_public.pdf

The input flows are the N fluxes (as tons of N emitted) from all other pools ('Energy and Fuels', 'Waste', 'Forest and semi-natural Vegetation', 'Materials and Products', 'Agriculture', 'Humans and Settlements', 'Hydrosphere') and N (reduced and oxidized) by transboundary transport. Output flows are N deposition to national ecosystems (hydro and terrestrial) and exported to neighbouring countries (Figure 1).

The pollutant emissions to be considered in this pool are NH_3, nitrogen oxides (NO_x) and N_2O. Even if properties of individual species may differ (e.g. N in NH_3, N in N_2O), N budgets refer only to total Nr.

NO_X is a generic term for mono-nitrogen oxides (NO and NO_2) and are mainly emitted during fuel combustion especially at high temperature. The main emitting sectors are industrial facilities and road transport.

The vast majority of NH_3 comes from the agricultural sector in connection with activities and practices such as manure storage, slurry spreading and the use of synthetic nitrogenous fertilizers.

N_2O is a powerful greenhouse gas produced both naturally and via human activities. N_2O gives rise to NO when reacting with oxygen atoms and this NO then reacts with ozone. It is the main naturally occurring regulator of stratospheric ozone (Ravishankara et al., 2009), by its depleting ozone activity. Over 100 years N_2O has a global warming potential (GWP) 298 times larger than carbon dioxide (CO_2) (IPCC, 2007). The main human sources of N_2O are agriculture, and especially soil cultivation with nitrogen fertilizers, and livestock production.

The NH_3, NO_X and N_2O input emissions to the atmosphere should be quantified from the output of the subpools 'Energy and Fuels', 'Agriculture' considering emissions from livestock and nitrogen fertilizers, 'Humans and Settlements' and 'Waste' sectors and natural emissions. Emissions are reported in tons of N emitted.

Inputs

Emissions

The principal source of the emission data has been the official Swedish reporting to the Centre on Emission Inventories and Projections (CEIP) under the Air convention (The Convention on Long-range Transboundary Air Pollution, CLRTAP). Since 2007 CEIP has been responsible for coordinating the emission-related work of EMEP. The reported NO_x and NH_3 emissions for the year 2015 are accessible at the CEIP webpage (www.ceip.at). The mandate of CEIP includes data quality assurance and the emissions reported by CEIP were used as Gridded aggregated NFR (Nomenclature For Reporting) sector data (GNFR). GNFR sector data were attributed to the individual N pools for six out of seven N pools (Tables 1 and 2). The exception was pool 6, (HS) "Humans and Settlements" with no attributable emissions according to the GNFR sector division. Emissions from GNFR sector N natural are a combination of emissions from two TFRN-defined pools; Forest and semi-natural vegetation, FS and Hydrosphere, HY. However, according to the CEIP database Sweden has reported zero natural emissions and therefore the emissions from natural vegetation, forests and from waters need to be quantified in an alternative way, based on parallel work on NNB pools 4 FS and 8 HY.

The Forest and semi-natural vegetation pool of the NNB (Jutterström et al., 2020) is divided into three different compartments: forest (FS.FO), wetland (FS.WL) and other land (FS.OL). For FS.FO and FS.WL the emissions of N_2, N_2O and NO to the atmosphere from denitrification and nitrification processes are included in the NNB. However, emissions from FS.OL to the atmosphere are considered negligible. Also, as N_2 is not reactive these emissions will not be added to the AT budget and will not be further discussed here. For 2015 the emissions of N_2O and NO from forest and N_2O from wetlands gave an addition to the atmosphere of 20.9 kt of reactive nitrogen. This also included emissions of N_2O due to land use and land-use changes (LULUC) for forest- and

wetlands (Swedish National Inventory Report, Statistics Sweden: http://www.statistikdatabasen.scb.se).

Emissions from HY were estimated to 0.5 kt (Stadmark et al., 2020). This is based on the calculated retention of 35 kt N of nitrogen deposited and emitted to inland waters before it reaches the sea and removal of nitrogen in coastal areas. The retention and removal are mainly due to denitrification and 1% of the denitrified N is assumed to enter the atmosphere as N_2O.

These emissions are in reality contributing to N deposition in Sweden. Even if these emissions are not included in the model they are accounted for at the step when the deposition from the MATCH model (Persson et al., 1994) is adjusted to agree with the deposition observed at monitoring stations. Natural emissions are also contributing to the Nr transported with air masses to outside the country.

Thus, the Swedish Nr emissions (Table 1) are dominated by the emissions from the pools AG (43.4 kt), EF (40.6 kt), MP (10.6 kt) and WS (1.0 kt), i.e. the sum of 95.6 kt Nr, according to CEIP reported emissions for the year 2015. The CEIP and the MATCH model, do not explicitly include natural emissions from FS (20.9 kt) and from HY (0.5 kt), together an additional 21.4 kt of Nr. The total anthropogenic and natural emissions are 117 kt.

Calculations of Nr deposition done by the MATCH Sweden system are based on the same emissions reporting as the EMEP model and despite the differences between the models, there is typically a high degree of consistency between EMEP and MATCH Sweden system outputs. In this report we used deposition based on the MATCH Sweden system (see below).

Table 1. Year 2015 Swedish emissions of NH_3 and NO_x to the atmosphere, based on the emissions submitted to CEIP (EF, MP, AG, WS and HS) and on our own estimates for FS and HY (Jutterström et al., 2020, Stadmark et al., 2020).

		N kt/yr
1 EF Energy and Fuels	**EF-AT**	40.6
2 MP Materials and Products	**MP-AT**	10.6
3 AG Agriculture	**AG-AT**	43.4
4 FS Forest and semi-natural Vegetation	**FS-AT**	20.9
5 WS Waste	**WS-AT**	1.0
6 HS Humans and Settlements	**HS-AT**	0
8 HY Hydrosphere	**HY-AT**	0.5

N from lightnings

The methodology to estimate emissions from lightning was found in Section 5 of chapter 11.C Other natural sources B111000 Lightning 2019 in the EMEP/EEA air pollutant emission inventory guidebook 2019 (EEA, 2019). Both cloud to ground lightning flashes as well as cloud to cloud flashes were considered. The emission factors needed for the calculations were given in section 8 and the statistics of lightning flashes in Sweden for 2015 was downloaded from the Swedish Meteorological and Hydrological Institute (https://www.smhi.se/data/utforskaren-oppna-data/blixtdata-historiska-arkivdata). For 2015 the number of cloud to ground flashes was close to 87 000 and there were approximately 63 000 cloud to cloud flashes. This resulted in a total contribution of 0.1 kt N. Note that about 20% of the emissions from cloud to ground emissions are assumed to be emitted below 1 km altitude and 80% below 5 km. The emissions from cloud to cloud flashes are assumed to be above 5 km altitude. In the budget, we have decided to include all the emissions regardless of emission height.

AT-N_2 – AT-N_r 0.1ktN

Outputs

Atmospheric nitrogen deposition

The reactive nitrogen deposition in 2015 in Sweden was 160 kt according to official national operational estimates by the MATCH Sweden system (Alpfjord and Andersson, 2017, Andersson et al., 2017, Andersson et al., 2018, Leung et al., 2019; https://www.smhi.se/data/miljo/atmosfarskemi), see Table 2. Most of this deposition originated from non-national emissions, 87% (Alpfjord and Andersson, 2017). Oxidised and reduced nitrogen species contribute equally (ca 50%) to the reactive nitrogen deposition in 2015. However, the national contribution (i.e. deposition in Sweden due to Swedish emissions) of reduced nitrogen deposition is a factor ca. 3 times higher than the corresponding national contribution of oxidised nitrogen deposition.

Table 2. Mean and total nitrogen deposition (oxidised, reduced and total reactive) in Sweden, Swedish contribution and non-national contribution (long-range transported). Estimates for the year 2015 by the MATCH Sweden system (operational annual re-analyses of nitrogen, sulfur, base cat ion, ozone deposition and air pollution concentrations) (Alpfjord and Andersson., 2017).

	Mean deposition to Sweden [mg N m^{-2} $year^{-1}$]			Total deposition [kt N $year^{-1}$]			Swedish total area (km^2)
Land use category	Oxidised nitrogen	Reduced nitrogen	Reactive nitrogen	Oxidised nitrogen	Reduced nitrogen	Reactive nitrogen	
Total (Sweden)	175	181	356	78.7	81.6	160	450000
Swedish contribution	10.5	33.6	44.1	5.00	16.0	21.0	
Long-range transport	164	148	312	73.7	65.6	139	

The MATCH Sweden system also estimates deposition to ecosystems, estimates for 2015 are included in Table 3. The Swedish weighted mean deposition is the ecosystem specific deposition flux, dependent on location, weighted by the grid-fractions of that ecosystem.

Table 3. Mean and total nitrogen deposition (oxidised, reduced and the sum reactive) into Swedish ecosystem types in 2015. Estimates for the year 2015 by the MATCH Sweden system (Alpfjord and Andersson, 2017), see also http://www.smhi.se.

Vegetation type	Swedish Area	Weighted mean deposition [mg N m^{-2} yr^{-1}]			Total deposition [kt N yr^{-1}]		
	Total [km^2]	Oxidised nitrogen	Reduced nitrogen	Reactive nitrogen	Oxidised nitrogen	Reduced nitrogen	Reactive nitrogen
Arable land	29 300	315	287	603	9.24	8.42	17.65
Pasture	16 900	259	245	504	4.37	4.13	8.50
Pine forest	144 900	124	141	265	17.93	20.41	38.34
Spruce forest	101 000	210	222	432	21.22	22.42	43.63
Deciduous forest	16 000	178	186	364	2.84	2.97	5.81
Lakes and Seas	80 700	202	211	412	16.27	17.00	33.27
Wetlands	32 100	107	110	217	3.45	3.53	6.99
Mountains	25 200	56	57	113	1.41	1.43	2.85
Beech and oak forest	1 400	644	512	1156	0.91	0.73	1.64

Atmospheric exports and imports

To establish the atmospheric pool of reactive nitrogen several simplifications have been made. The atmosphere pool is not divided into sub-pools, it only considers the troposphere and neglects fluxes of N between the different atmospheric layers. The loss of N from the atmosphere by N-fixation, either by natural processes or by fertilizer production using the Haber-Bosch process, is not considered in this pool since it - from the point of view of the atmosphere - only represents loss of N_2. Another simplification made is that the Nr from emissions outside Sweden which enters the country's air space through long-range transport is only considered if it gets deposited in Sweden. The part of long-range transport which enters the air space from a country upwind and gets transported to the country downwind is not quantified. Consequently, the long-range export of Nr originates only from the Swedish Nr emissions. In this work we assume that the atmospheric Nr pool is constant. Therefore, the long-range export is calculated as a difference between known inputs (import, emissions, lightnings) and outputs (deposition). Finally, the emissions from Forest and semi-natural vegetation and from Hydrosphere are not considered in the MATCH model.

Modelled deposition, however, includes the deposition from natural emissions as the model estimates are analysed and assimilated together with measured data/observations that include deposition from both natural and anthropogenic sources. In summary, the inputs to the atmospheric pool of Nr are long-range transport, Swedish emissions (all sectors + natural emissions) and Nr production by lightnings. The outputs are Nr deposition and long-range transport of Nr to countries downwind. These major inputs and outputs are summarised in Figure 2.

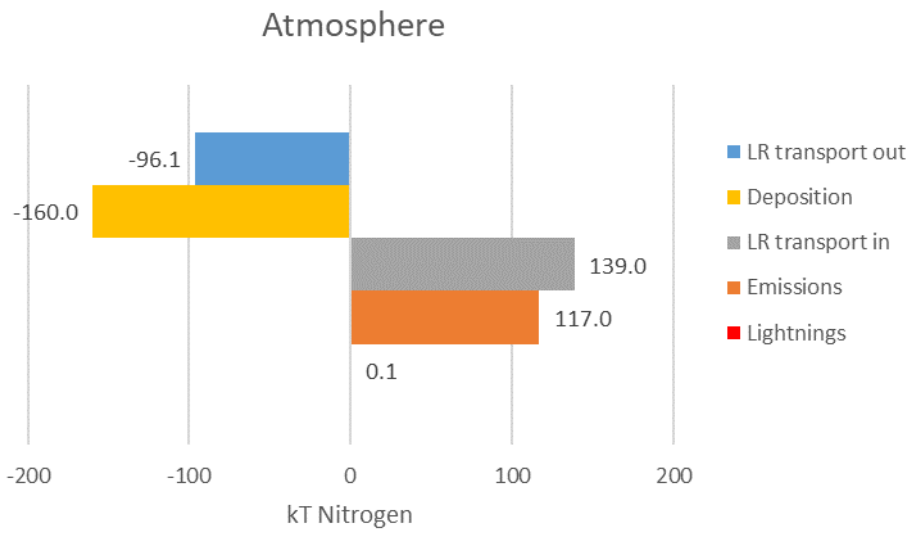

Figure 2. Annual loss of Nr from the atmospheric pool of Nr is mainly by two processes: by the transport of emitted Nr with air masses to outside Swedish territories and by deposition of Nr on land and water within Sweden. The inputs are transboundary air pollution imported with air masses from sources outside Sweden, emissions of Nr in Sweden and Nr produced by lightnings. Units: kt N/year, all calculations for year 2015.

Alpfjord and Andersson (2017) calculated that 76% of the Swedish anthropogenic nitrogen emissions are exported from Sweden and deposited elsewhere. In this report we have used the same deposition calculations and updated emission data from CEIP. The overall picture of the main Nr flows to and from troposphere above the geographical area of Sweden for the year 2015 is provided by Figure 3. As opposed to several other pools such as e.g. Agriculture, the atmospheric part of the NNB consist of only one pool (the atmosphere) with no sub-pools. The work behind mapping and reporting the national emissions and the modelling and monitoring work behind calculating the transboundary movement of the Nr is in both cases extensive and involves many expert teams and considerable resources. However, this work is carried out on a regular basis as a part of national reporting to LRTAP and UNFCCC and the data compilation presented here is on the emission side based on public data accessible online.

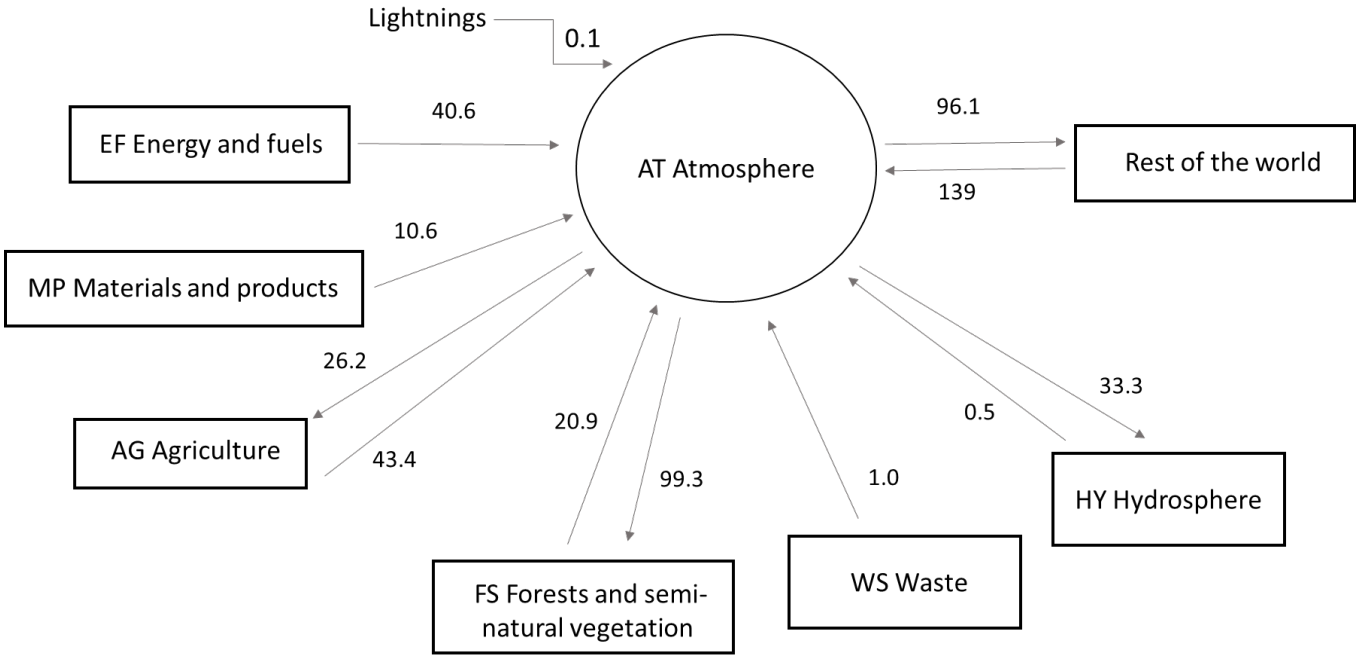

Figure 3. Main flows to and from the atmospheric pool of reactive N. Units: kt N, year 2015

Conclusions

Major flows to and from the atmospheric pool of Nr were calculated for the year 2015 for the air masses of the troposphere above Sweden, including the Swedish national coastal waters. With exception of the small amount of Nr generated by lightnings, the atmospheric Nr pool is dominated by four main flows: import and export of Nr with air masses entering and leaving Swedish air space, by Swedish emissions of Nr and by atmospheric deposition. Sweden is a net importer of air pollution in the form of Nr, i.e. the import of Nr from the emission sources outside the country (139 kt) is larger than the export to countries downwind (96.1 kt). The deposition of Nr originates by 87% from emission sources abroad while the emissions of Nr generated in Sweden are to 82% transported outside Sweden. The largest emissions come from two sectors: Agriculture (43.4 kt) and Energy & Fuels (40.6 kt), which together stand for 72% of the Swedish emissions. The atmospheric pool of NNB is one of the 8 major pools defined by the TFRN which together represent a total national nitrogen budget capturing all major flows of all forms of Nr within a country and across the country borders.

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