At the boundary between anthropogenic and environmental systems: the neglected emissions of indirect nitrous oxide

Arnaud Helias

At the boundary between anthropogenic and environmental systems: the neglected emissions of indirect nitrous oxide.

Arnaud Hélias[1, 2, 3]

[1]LBE, INRA, Montpellier SupAgro, Univ Montpellier,11100, Narbonne, France;

[2]Elsa, Research group for Environmental Lifecycle and Sustainability Assessment, 34060, Montpellier, France;

[3]Sustainable Engineering, Technische Universität Berlin, Berlin, Germany.

Corresponding author: Arnaud Hélias
Address : Montpellier SupAgro, 2 place Pierre Viala, 34060 Montpellier Cedex 1, France
E-mail: arnaud.helias@supagro.fr; Phone : +334 99 61 27 65; FAX : no fax number

1. Introduction

Nitrous oxide (N_2O) is the third most important Greenhouse Gas (GHG) after carbon dioxide and methane. Its emissions must be quantified and even more so for agricultural activities, which are the main human interventions in the global nitrogen cycle. This paper focuses on how Life Cycle Assessment (LCA) deals with this gas. It discusses what is included in the inventory of activities and what is included in the impact assessment and shows the inconsistency of modelling between agricultural and non-agricultural activities.

To address this topic, it is necessary to begin by presenting the general structure of the assessment. The availability of a simple carbon footprint measurement has become one of the key tools for stakeholders to develop awareness of climate change issues. Due to the complexity of the mechanisms relating human activities and climate change, the modelling strategy can be planned in two steps: first, the "carbon footprint practitioners" model anthropogenic activities in order to determine GHG emissions and second, the "climate scientists" model global warming mechanisms in order to provide the Global Warming Potential (GWP). On one hand, GHG emissions result from human activities and need to be assessed for each studied alternative or system. On the other hand, GWPs are global, independent from the activities and only change according to science updates, with International Panel for Climate Change (IPCC) assessment report editions. In other words, carbon footprints are the results of GHG × GWP. Only GHGs vary according to the anthropogenic systems whereas GWPs remain constant for the practitioners, providing the environmental system is assumed to be stable from one study to another.

This understanding of the carbon footprint matches with the LCA framework in which the system is divided into two steps: First, the Life Cycle Inventory (LCI) describes emissions from techno-economic activities (anthropogenic system) towards the soil, water and air compartments (the environmental system). Second, the Life Cycle Impact Assessment (LCIA) converts elementary flows into impacts and damages using Characterization Factors (CFs). LCA practitioners perform LCA by producing the LCI and by using CFs from LCIA (LCA = LCI × CFs) while the LCIA scientists provide CFs by modelling the causal fluxes into the environment.

This partitioning is useful, and seems both obvious and simple. LCA practitioners assess the environmental issues of products and services without exhaustive knowledge and ad hoc modelling of environmental mechanisms. For the interpretation of results, it is mandatory to understand the general structure and the main hypotheses of LCIA methods. However, when an LCA is executed, the environmental modelling task is simplified by using CFs within the LCA software.

Nonetheless, this partitioning can also be uncertain; although the limit between LCI and LCIA is most often obvious, sometimes a sequence of steps can occur, making it difficult to define the boundary between them. This has previously been discussed for pesticides

(Rosenbaum et al. 2015) and spatial and temporal limits have thus been proposed (van Zelm et al. 2014). Weidema et al. (2017) provide the outline of the delimitation between LCI and LCIA and they suggest how this dichotomy could be overcome. In the present paper, the significance of N_2O emissions and their place in LCA are discussed.

2. Methods

2.1. Direct and indirect N_2O emissions

N_2O releases are thoroughly addressed in the agricultural section of the IPCC report (De Klein et al. 2006), differentiating between direct and indirect nitrous oxide emissions (in the LCA community, the latter are sometimes called induced emissions while the "indirect" term is rather employed for background emissions). Direct emissions merge few atmospheric N_2O releases, with air emissions resulting in N that enters managed (agricultural) soil. Indirect emissions result from re-deposition after volatilization and leaching/runoff of other N compounds, essentially ammonia (NH_3), NOx and nitrate (NO_3^-). These induce secondary emission flows.

Fig. 1 describes the impact pathway, linking N emissions from human activities to global warming. This illustrates the relationships between anthropogenic and environmental systems. The involved mechanisms include:

1. Direct N_2O releases. These emissions are obviously driven by the anthropogenic system, which is determined by technological or functioning choices. This flow has to be quantified in the LCI.
2. Other direct N-compound emissions. They also have to be quantified in the LCI.
3. Emissions of N_2O and other N-compounds after N incorporation into the soil, considered as direct. These emissions depend upon the spreading quantity and their nature (chemical composition), however they are also driven by the

environmental system, such as pedoclimate conditions, rainfall or atmospheric temperature. These aspects constitute pieces of information for spreading management. They are highly specific and have to be adapted to each condition. Direct emissions are part of the practitioner's task and contribute to LCI work. From this standpoint, agricultural soil is considered as a part of the technosphere (i.e. part of the production system).

4. Mechanisms for re-deposition of other N-compounds and/or for their biological conversion to N_2O. These mechanisms are unrelated to the product described. They are sensitive to climatic conditions and to spatial variations but with too wide a scale to be specific to a given study. They are only driven by the environmental system. However, these indirect emissions need to be modelled by the practitioner, because CFs for climate change (GWP) only represent the radiative forcing and the degradation of the molecules in the atmosphere. These environmentally driven mechanisms enter the LCI since they are not represented by the LCIA.
5. The conversion of the mass of a molecule into its impact on global warming is obviously part of the LCIA (this also concerns the later steps of methods that reach endpoint damage).

Insert Figure 1 about here

2.2. Modelling of N_2O emissions

Covering the different carbon footprints and emission factor databases, models are employed for determining direct N_2O emissions (flows 1, 2 and 3 to N_2O in Fig 1). Models originate from various sources: IPCC tier 1, country level factors, or site-specific data. Concerning the releases of other N-compounds (flows 3 to NH_3, NO_x and NO_3^- in Fig. 1), many approaches are used such as direct measurements, site-specific models or international guidelines like IPCC tier 1 or other similar sources for ammonia (2016) and nitrates (Roy et al. 2003).

For indirect emissions (flow 4), LCAs (and carbon footprints) only use one model, described in the tier 1 IPCC guidelines: this involves 1% of volatilized and 0.75% of leached N-compounds. All works use the same conversion factors in the LCI to represent this environmental mechanism of re-deposition and/or biological conversion, which are not influenced by the technical system. This is well documented for agricultural sectors, which are the main contributors of N_2O emissions. Practitioners commonly calculate direct and indirect emissions for agricultural products but this raises an issue for non-agricultural sectors.

2.3. Difference between agricultural and non-agricultural activity sectors

2.3.1. Guidelines for state-level assessments

IPCC guidelines, Chapter 11, stipulate that “the sources of N as NH_3 and NO_x are not confined to agricultural fertilisers and manures, but also include fossil fuel combustion, biomass burning, and processes in the chemical industry” (De Klein et al. 2006). Moreover, in Chapter 7 they specify that “it is good practice to estimate and report N_2O emissions from atmospheric deposition of NO_x and NH_3 where a country already has an inventory of these gases” (Gillenwater et al. 2006).

These considerations have been quoted in a national emissions report guideline where “Parties may report, as a memo item, indirect N_2O emissions from other than the agriculture and LULUCF [Land use, land-use change, and forestry] sources. These estimates of indirect N_2O should not be included in national totals. For Parties that decide to report indirect CO_2, the national totals shall be presented with and without indirect CO_2.” (UNFCCC 2014).

This points to a compromise between theoretical (non-agricultural indirect N_2O emissions should be reported), and operational assessments (these indirect emissions are often non-assessable and generally low). They are neglected to allow comparisons to be made with a same perimeter for all parties: both direct and indirect emissions are used for agricultural activities, while other activities only involve direct emissions.

2.3.2. Product carbon footprint and LCA studies

The carbon footprints of products are estimated in the same way as for countries. Ammonia emissions from industries or nitrates from wastewater treatment plants do not lead to any consequence on climate change, while crop production and animal waste spreading do have effects. Indirect N_2O emissions do not result from NO_x from combustion, while they are a product of N field fertilisation. Since it is not mandatory in the current guidelines, carbon footprint practitioners do not deal with these non-agricultural emissions (some are probably quantified but this remains an exception).

This differs for LCA practitioners because N-compounds lead to other impacts. Regarding the latest recommendation for the European product environmental footprint (Fazio et al. 2018), NH_3, NO_3^- and NO_x are involved in marine and terrestrial eutrophication, NH_3 and NO_x in acidification and particulate matter, and NO_x in photochemical ozone formation. Emissions of these substances are quantified in LCI for all activity sectors. They lead to these impacts through the characterization factors of LCIA methods. Through this inventory work for other impacts, it is possible to quantify indirect N_2O emissions for all activity sectors.

2.4. Quantification of indirect N_2O emissions for non-agricultural sectors

This quantification is performed by providing CFs to represent the indirect N_2O emissions. They are applied to the datasets in the ecoinvent 3.4 database (Wernet et al. 2016), with an "at point of substitution" system model. These CFs are obviously not used with the agricultural datasets as the mechanism has already been taken into account at LCI level.

The CFs to assess GWP of N-compounds through indirect N_2O emissions are calculated from their N-content, the re-deposition (1%) and leaching (0.75%) of indirect emission factors according to the emission compartment, and the IPCC2013 N_2O GWP_{100y} (265 kg_{CO2eq}/kg_{N2O}); see Table 1. In addition to NH_3, NO_3^- and NO_x, the closely related chemical forms (NH_4^+, NO_2^-, NO) and unspecific emissions (nitrogen, and organic bound nitrogen) are also taken into account.

Insert Table 1 about here

3. Results

Firstly, the CFs in Table 1 are used for assessing the climate change impacts of each unitary dataset (i.e. unit process). This implies the impact of its elementary flows, without determining the process tree and the impact associated to its technology inputs. Out of the 14 927 datasets of the ecoinvent 3.4 database, 9 070 do not involve global warming effects nor N-compound elementary flows. These include market processes without any elementary flows but only technical ones.

The variation of climate change impacts for the remaining 5 857 is illustrated in Figure 2. In these sets, 1 907 show no change in impact. For these datasets, which are agricultural or do not contain any N compounds (387), ecoinvent already computes the indirect effect in the LCI. 67% remain, that show a variation; for 19% of processes, the increase is greater than 5%; for 16% it is greater than 10% and for 7%, greater than 100% (this latter group contains the 334 processes without conventional climate change impact but with N-compounds). If each ecoinvent entry is considered separatly, indirect N_2O emissions from N-compounds cannot be neglected for the non-agricultural sectors. Indeed, the relative variation is too high.

Insert Figure 2 about here

Figure 3 illustrates the result with cumulative datasets (with the impact of the whole process tree) where the lack of indirect emissions is less of an issue. Over all the ecoinvent 3.4 datasets, 219 do not have global warming effects, nor N-compound elementary flows. Over the remaining 14708 datasets, the impacts change for almost all of them (99%) although they remain mostly small. For only 11%, the increase exceeds 1%, while for just 157 datasets (1%), the increase is greater than 5%. Nevertheless for this small fraction of datasets, climate change due to N-compounds cannot be neglected. Data from Figure 2 and 3 are available in the supplementary materials section of this article.

Insert Figure 3 about here

4. Discussion and conclusion

4.1. Current situation: inconsistency between LCI and LCIA

There is a noteworthy LCI/LCIA dichotomy in LCA. On one hand, for LCI, the result is "the quantitative description of flows of matter, energy, and pollutants that cross the system boundary"(Jolliet et al. 2016). On the other hand, for LCIA, "impact pathways consist of linked environmental processes, and they express the causal chain of subsequent effects originating from an emission or extraction."(Jolliet et al. 2004). Continuity across the border between these two types of result is essential.

The current climate change impact assessment results from its history, evolving from the first GWP values at the beginning of the nineties to present-day guidelines and pathways. This implies that the environmental system (the LCIA) starts with the radiative forcing of GHG. To ensure continuity, the environmentally driven indirect N_2O emissions should be described in the anthropogenic system (the LCI), however for sakes of simplicity, this is only done for the agricultural sector.

The final result depicts an inconsistency between LCI and LCIA for the non-agricultural sectors. Fortunately, for most of the studies, these missing emissions do not represent a strong contribution to climate change, but they do matter in some cases. It is more of an issue at unit process levels.

4.2. Proposal of a new boundary

The representation of environmental mechanisms in LCI is supported by an environmental context affecting human decisions (and associated emissions) or a specific situation which cannot be represented through a simple CF[1]. Nonetheless, this is not true for re-deposition of N-containing compounds and/or biological conversion to N_2O. These mechanisms do not contribute to the assessment of initial emissions. All practitioners use the same representation provided by IPCC.

The system border between LCI and LCIA should be movable and this should be easy to achieve. The indirect emissions from N-Compounds (flow 4 in Fig. 1) would be included in the LCIA by using the CFs listed in Table 1 and the indirect emissions would then be removed from agricultural datasets. That can be easily automated to update a database like ecoinvent.

---

[1] This will be less and less tenable with spatialized and temporalized CFs, which are progressing for many kinds of impact, as has been done for water since several years.

In the case where a practitioner does not agree with the IPCC indirect emission factors, he could investigate further into these mechanisms and propose improved CFs. These latter values would then be available to all practitioners. Representing redeposit and leaching in the LCIA would make it possible to introduce spatial considerations through regionalized CFs, involving more complex mechanisms such as atmospheric circulation, soil properties and soil cover (see for example Bühlmann et al. (2015) for the latter). Regionalized CFs would also allow for the consideration of country-level guidelines.

With this pragmatic boundary, by assigning the anthropogenic system to LCI and the environmental system to LCIA, direct emissions (flows 1 and 2 for non-agricultural activities, and flow 3 for agricultural spreading activities in Figure 1) remain in the LCI. This is obvious for 1 and 2 which are only driven by human activities. Flow 3 results from both systems: a spreading operation decided by a farmer, partially based on environmental considerations. To represents this specificity, flow 3 has to remain in LCI. However in the latter case, a default model of emissions subsequent to soil N-input could be used to determine CFs when LCA practitioners do not have access to extensive information on spreading conditions. This has been done for marine eutrophication in the 2008 Recipe version (Goedkoop et al. 2013). For this, CFs were available for elementary flows of “fertilizer, applied (N-component)” and “manure, applied (N component)” (names are those used in Simapro software) to represent volatilization and leaching mechanisms, and for the rest of the impact causal chain. This has not been done elsewhere and is not included in the 2016 Recipe version anymore, as N-limited eutrophication is no longer assessed.

4.3. Challenges for the new boundary

From a technical point of view, the new LCI/LCIA boundary does not raise any difficulties, but global warming is the most assessed impact in LCA and concerns a much broader scope: a change in the way it is addressed should be considered with caution.

With indirect N_2O emissions in LCIA, the N-compounds involved come up as “new” GHGs, in addition to the IPCC list. This should therefore be accompanied by the necessary explanations, showing the consistency between these new factors and the mechanisms previously described by the IPCC. It will also be necessary to explain that ignoring indirect emissions for non-agricultural sectors can no longer be justified, since precursors must be quantified for other impacts. Any change in the way global warming is assessed cannot be made without significant communication efforts.

Changing the LCI/LCIA boundary requires ensuring consistency between LCI-level work and LCIA uses. For the agricultural sector, LCIs with indirect N_2O emissions (old LCIs that are not updated or new datasets but defined as previously) must not be assessed with the new CFs to avoid double counting. The opposite situation is also true. New LCIs (without redeposit and leaching/runoff) must not be associated with current LCIA method, to avoid neglecting indirect emissions.

Since agricultural LCA-practitioners are aware of indirect N_2O emissions, these kind of issues should not be too prevalent. Nevertheless, if the definition of a new LCI/LCIA boundary allows a better articulation between these two steps, it remains for practitioners to ensure the consistency of the modelling.

4.4. Alternative and concluding remarks

An alternative is to keep indirect N2O emissions at the LCI level and add them for non-agricultural datasets in the LCI databases. This could also be easily automated by database providers, by adding a part of N-compound emissions (determined by IPCC indirect emission factors) to direct N_2O emissions.

In this scenario, no change is required at the LCIA level and the LCI/LCIA boundary is not modified. It is easy to set up. However, two points must be raised: (1) LCA practitioners modelling new non-agricultural inventories must change their habits to take into account the indirect emission of N_2O, which could be a challenge. (2) The environmental mechanisms of indirect emissions remain at the LCI level, which does not fit to the implicit separation of anthropological and environmental systems in the LCI/LCIA framework. That means that future improvements in environmental mechanism modelling, with spatialized re-deposition and more accurate re-emission models, will have to be implemented by practitioners and the LCI databases updated. A new LCI/LCIA boundary seems more relevant: systems with human intervention in LCI and environmental systems without human involvement in LCIA.

More generally, impact pathways related to nitrogen compounds should be harmonised, as they involve the same redeposit mechanisms as for climate change, terrestrial acidification and eutrophication. The same model and identical boundaries with LCI should be used in the LCIA methods.

CFs are increasingly accurate regarding environmental mechanisms. The quality and quantity of inventory datasets are also improving rapidly. It is therefore crucial to ensure a consistency between the two LCA mainstays.

5. Acknowledgments

The author thanks his colleagues A. Benoist, C. Bessou, L. Lardon and P. Roux for the fruitful discussions that led to this article. He is grateful to the ecoinvent Centre for providing the unit version of the database. Anonymous reviewer is warmly thanked for his valuable comments that helped improving this paper.

References

Bühlmann T, Hiltbrunner E, Körner C, et al (2015) Induction of indirect N_2O and NO emissions by atmospheric nitrogen deposition in (semi-)natural ecosystems in Switzerland. Atmos Environ 103:94–101. doi: 10.1016/j.atmosenv.2014.12.037

De Klein C, Novoa RSA, Ogle S, et al (2006) Chapter 11. N2O Emissions From Managed Soils, and CO2 Emissions From Lime and Urea application. In: 2006 IPCC Guidelines for National Greenhouse Gas Inventories. p 11.1-11.54

Fazio S, Castellani V, Sala S, et al (2018) Supporting information to the characterisation factors of recommended EF Life Cycle Impact Assessment method. EUR 28888 EN, European Commission, JRC109369, Ispra

Gillenwater M, Saarinen K, Ajavon A-LN (2006) Chapter 7. Precursors and Indirect Emissions. In: 2006 IPCC Guidelines for National Greenhouse Gas Inventories. p 7.1-7.16

Goedkoop M, Heijungs R, Huijbregts M, et al (2013) ReCiPe 2008 - First edition (revised) - Report I: Characterisation. The Hague, Netherland

Jolliet O, Müller-Wenk R, Bare J, et al (2004) The LCIA midpoint-damage framework of the UNEP/SETAC life cycle initiative. Int J Life Cycle Assess 9:394–404. doi: 10.1065/lca2004.09.175

Jolliet O, Saadé-Sbeih M, Shaked S, et al (2016) Environmental Life Cycle Assessment. Taylor & Francis

Rosenbaum RK, Anton A, Bengoa X, et al (2015) The Glasgow consensus on the delineation between pesticide emission inventory and impact assessment for LCA.

Int J Life Cycle Assess 20:765–776. doi: 10.1007/s11367-015-0871-1

Roy RN, Misra RV, Lesschen JP, Smaling EM (2003) Assessment of soil nutrient balance. FAO Fertil plant Nutr Bull 14:101p

UNFCCC (2014) Report of the Conference of the Parties on its nineteenth session; held in Warsaw from 11 to 23 November 2013; Addendum; Part two: Action taken by the Conference of the Parties at its nineteenth session FCCC/CP/2013/10/Add.3

van Zelm R, Larrey-Lassalle P, Roux P (2014) Bridging the gap between life cycle inventory and impact assessment for toxicological assessments of pesticides used in crop production. Chemosphere 100:175–181. doi: 10.1016/j.chemosphere.2013.11.037

Weidema BP, Schmidt J, Fantke P, Pauliuk S (2018) On the boundary between economy and environment in life cycle assessment. Int J Life Cycle Assess 23:1839–1846. doi: 10.1007/s11367-017-1398-4

Wernet G, Bauer C, Steubing B, et al (2016) The ecoinvent database version 3 (part I): overview and methodology. Int J Life Cycle Assess 21:1218–1230. doi: 10.1007/s11367-016-1087-8

(2016) EMEP/EEA air pollutant emission inventory guidebook 2016 - Technical guidance to prepare national emission inventories. European Environment Agency, Luxembourg

Table 1. Characterization factors of N-compounds

N-Compound[1]	N-content (%)	Emission compartment	Indirect emission factor (%)	CF (kg_{CO2eq}/kg)
Ammonia (NH_3)	82%	air	1%	2.18
Ammonium carbonate ($(NH_4)_2CO_3$)	29%	air	1%	0.77
Ammonium, ion (NH_4^+)	78%	water	0.75%	1.54
Nitrate (NO_3^-)	23%	air	1%	0.60
Nitrate (NO_3^-)	23%	soil, water	0.75%	0.45
Nitric oxide (NO)	47%	air	1%	1.24
Nitrite (NO_2^-)	30%	water	0.75%	0.61
Nitrogen (N)	100%	air	1%	2.65
Nitrogen (N)	100%	soil	0.75%	1.99
Nitrogen oxides (NO_x)	30%[2]	air	1%	0.81
Nitrogen, organic bound (N)	100%	water	0.75%	1.99

[1] Ecoinvent elementary flow names.

[2] Assuming a molar mass of NO_x equals to NO_2.

Figure captions

Figure 1. Schematic of the pathways involved in N_2O release. 1–3: direct emissions, 4: indirect emissions, 5: impact assessment. See text for details.

Figure 2. Relative increase of climate change impacts of unitary datasets by taking into account indirect N_2O emissions for all activity sectors.

Figure 3. Relative increase of climate change impacts of cumulative datasets by the consideration of indirect N_2O emissions for all activity sectors.

Figure 1

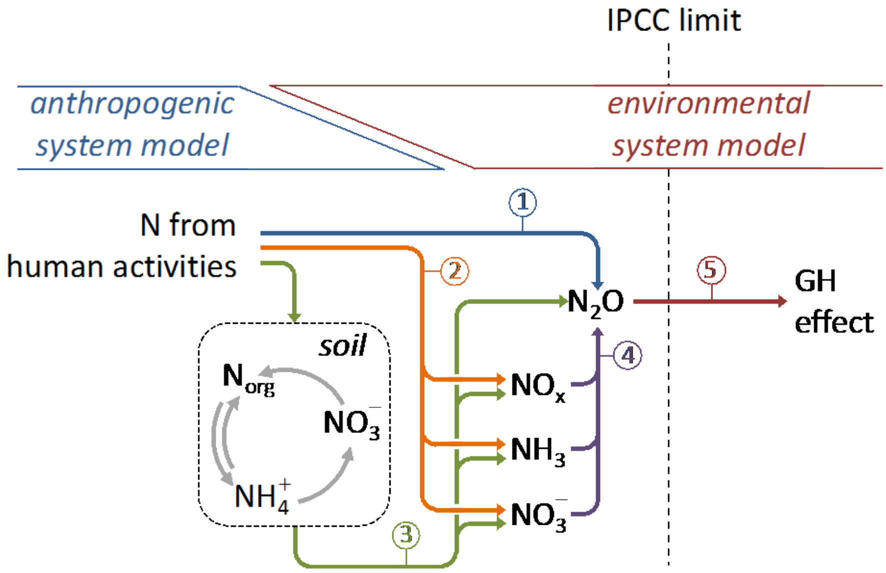

IPCC limit
anthropogenic system model
environmental system model
N from human activities
soil
N_{org}
NO_3^-
NH_4^+
①
②
③
④
⑤
N_2O
NO_x
NH_3
NO_3^-
GH effect

Figure 2

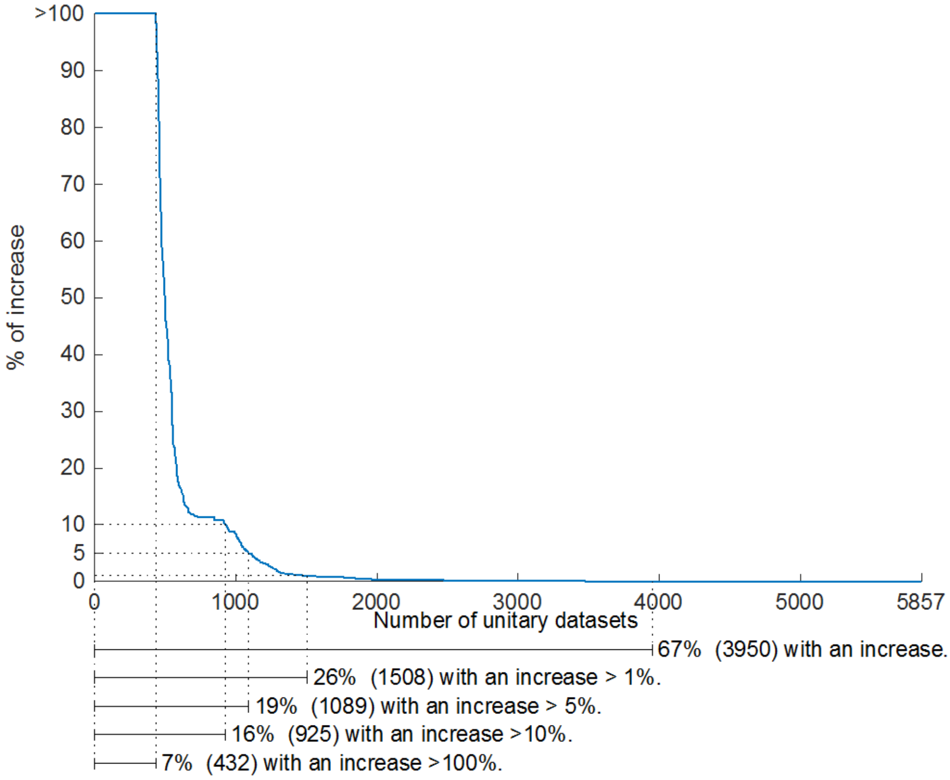
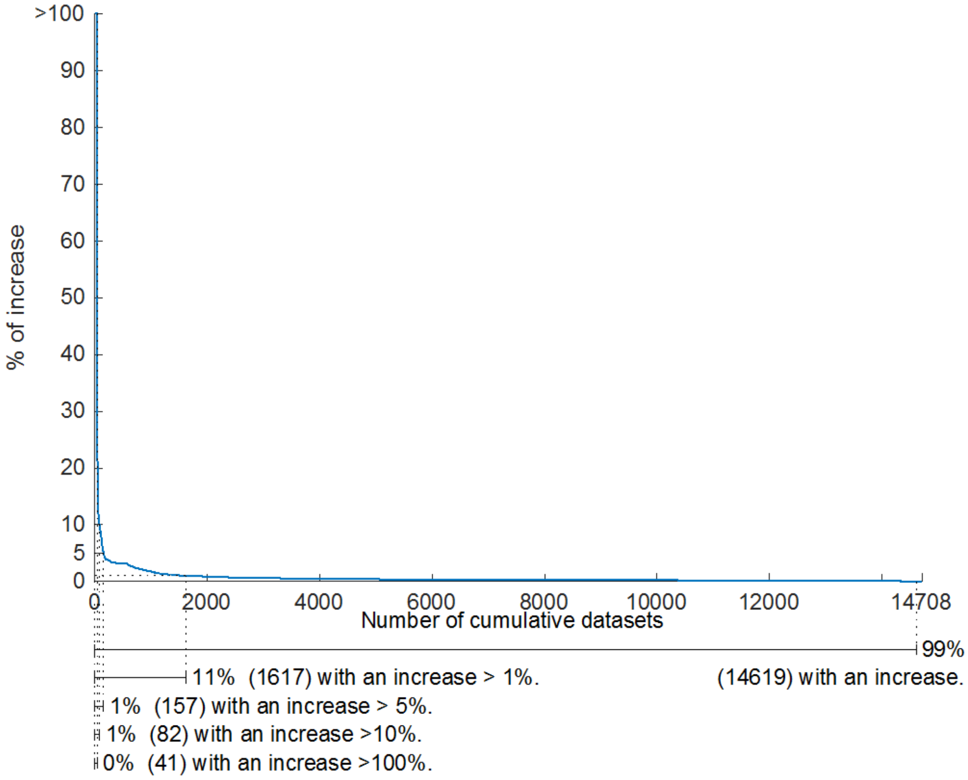

% of increase
>100
90
80
70
60
50
40
30
20
10
5
0
0
1000
2000
3000
4000
5000
5857
Number of unitary datasets
67% (3950) with an increase.
26% (1508) with an increase > 1%.
19% (1089) with an increase > 5%.
16% (925) with an increase >10%.
7% (432) with an increase >100%.

Figure 3


% of increase
>100
90
80
70
60
50
40
30
20
10
5
0
0
2000
4000
6000
8000
10000
12000
14708
Number of cumulative datasets
99%
(14619) with an increase.
11% (1617) with an increase > 1%.
1% (157) with an increase > 5%.
1% (82) with an increase >10%.
0% (41) with an increase >100%.