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Testing persistence of ammonia emissions with historical data from 1770 to 2019 in 37 OECD countries

Solarin Sakiru
University of Nottingham
Lorenzo Bermejo
Universidad Villanueva
Luis A. Gil-Alana (alana@unav.es)
University of Navarra

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Abstract

We examine the historical time series data of ammonia emissions from 1770 to 2019 in 37 OECD countries by looking at its statistical properties in order to determine if the series display time trends and persistence. These two properties are very common in environmental data, and our results indicate that reversion to the mean only occurs in the case of Finland, while the null hypothesis of a unit root cannot be rejected in the case of Norway or Iceland. In all the other cases, the estimated value of the differencing parameter is much higher than 1, and this is consistent for the two assumptions made regarding the error term. Thus, shocks are expected to be permanent in all cases except Finland.

1. Introduction

NH_3 (ammonia) is the most abundant alkaline gas in the atmosphere, it is a highly reactive and soluble alkaline gas, which originates from both natural and anthropogenic sources. Ammonia comes from the decomposition and volatilization of urea. High-density, intensive agricultural practices are considered "hot spots of emission." Ammonia emissions related to agriculture, such as the burning of biomass or the manufacture of fertilizers, are also relevant. Other sources of NH_3 emissions come from catalytic converters in gasoline fuelled cars, landfills, sewers, composting of organic materials, combustion, industry, birds and wild animals and volatilization from soils and oceans (Sutton et al., 2000; Bicer et al., 2017).

Recent studies indicate that NH_3 emissions have increased worldwide in recent decades. Ammonia has impacts both locally and internationally. In the atmosphere, ammonia reacts with acidic pollutants such as the products of NOx and SO_2 emissions to produce a fine aerosol containing ammonia (NH_4+). In this sense, although the useful life of NH_3 is relatively short (< 10–100 km), NH_4 + can be transferred over longer distances (100-> 1000 km) (Fowler et al., 1998; Asman et al., 1998; etc.). This is a serious problem, since NH_3 plays a very important role in the formation of atmospheric particles, the degradation of visibility and the atmospheric deposition of nitrogen in sensitive ecosystems. Excess nitrogen may cause eutrophication and acidification effects in semi-natural ecosystems, which in turn may lead to species composition changes and other deleterious effects (Van den Berg et al., 2008; Wiedermann et al., 2009; Bobbink et al., 2010; etc.). In short, the increase in NH_3 emissions has a high negative impact on public and environmental health and, without a doubt, on climate change (Behera et al., 2013; Lourenço & Nunes, 2020).

One of the features of ammonia emissions which has not been examined in the existing literature is its persistence. Persistence of ammonia emissions suggests that shocks to ammonia emissions in these countries will have permanent effects. The magnitude of the persistence of ammonia emissions will determine the size of the remedial measures required by the authorities to confront the harmful impact associated with any sudden increase in ammonia emissions. Furthermore, persistent ammonia emissions suggests that an abrupt increase in ammonia emissions (probably due to industrial discharges, changes in manure application, agricultural and urban runoff, drainage from fish and shrimp farms) will continue into the future unless drastic actions are taken by policy makers to restrain such an increase. Persistence of ammonia emissions suggests that it is difficult to correctly project future values of ammonia emissions by only relying on its past trend. Additionally, if two or more ammonia emissions series are stationary, the prospect of convergence between the series is very lean and accordingly it might be incorrect to make a convergence inference on such series (Nieswiadomy & Strazicich, 2004).

We examine historical time series data referring to the ammonia emissions in 37 OECD countries starting in 1770 and ending in 2019 in this paper. Ammonia emissions grew in OECD members in most of the years covered in this study. For example, ammonia emissions expanded in OECD countries from 847 kilotons in 1770 to 2,710 kilotons, 11,320 kilotons, and 11,601 kilotons in 1900, 1990 and 2019, respectively (Feng et al., 2020). We focus on issues such as the existence of deterministic terms and persistence which are both features widely observed in environmental studies (Gil-Alana et al., 2017; Zhang et al., 2020; Solarin et al., 2021). Our results indicate that time trends are statistically significantly positive in six countries (Turkey, Australia, Canada, New Zeeland, Norway and Iceland) independently of the specification of the error term, but also in Mexico, Spain, Italy, Chile, Austria and Slovenia if the errors in the differenced process are uncorrelated. On the other hand, mean reversion, and thus, transitory shocks, are only observed in the case of Iceland. The unit root hypothesis cannot be rejected for Norway and Iceland, and for the remaining countries the degree of differentiation seems to be significantly higher than 1.

The rest of the paper is structured as follows: Section 2 presents a short review on the literature on modelling environmental data; Section 3 describes the dataset and the methodology used based on the concept of fractional integration. Section 4 is devoted to the empirical results, while Section 5 concludes the paper.

2. Literature Review

With the increase in population, the need to generate enough food to meet this growth has also raised. Fritz Haber achieved, at the beginning of the 20th century, the synthesis of NH_3. The process consisted, basically, of converting inert gaseous N_2 into biologically active forms that were used to fertilize fields and increase food production, which made it possible to meet the demand of considerable population increases, as reported in the works by Erisman et al. (2007) and Reis et al. (2009) and others. But this beneficial effect resulted in the addition of an excess of anthropogenic nitrogen (N) compounds to the atmosphere. This substantial increase has become a major problem and concern for human health and the environment, as stated by Krupa and Moncrief (2002) and Behera et al. (2013) among many others. The most important N gases that are emitted by human activities are nitrogen oxides (NOx), nitrous oxide (N_2O) and NH_3. From these gases, NH_3, is emitted, as explained by Olivier et al. (1998) and Zhang et al. (2008), by a large number of sources, such as the volatilization of animal waste and synthetic fertilizers, loss of soil under native vegetation and agricultural crops, human excrement and combustion of fossil fuels.

The existence of NH_3 in the gaseous phase and its interaction with other substances in the atmosphere was discovered in the last century. Being the only kind of primary alkaline basic gas in the atmosphere, NH_3 plays, as Shukla and Sharma (2010), Xue et al. (2011) and Behera et al. (2013) argue, an important role in determining the general acidity of precipitation, airborne particles (aerosols and PM) and cloud water. Ammonia and ammonium (NHx) are also nutrients that fertilize plants, as reflected in the work of Sutton et al. (2000) and Alonso et al. (2017). However, a considerable increase in the anthropogenic contribution of N to the environment can lead to the eutrophication of terrestrial and aquatic ecosystems, which poses a serious threat to biodiversity (see, e.g., Asman et al., 1998; Galloway et al., 2003; Erisman et al., 2005; Van den Berg et al., 2008; Wiedermann et al., 2009; Bobbink et al., 2010; Wang et al., 2022).

More recently, studies such as Charlson et al. (1990), Bauer et al. (2007), Myhre et al. (2009) and Lourenço & Nunes (2020) have examined the impact of the sources, the movement and destination of atmospheric NH_3 on climate change that has been taking place worldwide. NH_3 emissions have increased worldwide in recent decades, due to atmospheric ammonia having impacts both locally and internationally as shown in the studies by Asman et al. (1998) and Fowler et al. (1998). Specifically, the effects of sulphate (SO_4^{2-}) and nitrate (NO_{3-}) aerosols on the dispersion of incoming solar radiation have been verified. The greater the availability of aerosol particles, the greater the cloud droplet formation. As a consequence, the total accumulated area of all the droplets is larger, the resulting cloud is more reflective and remains longer (cloud life effect).

In summary, ammonia is a nitrogen-containing compound and its emissions contribute to the formation of ammonium sulphate and ammonium nitrate aerosols, which deteriorate air quality. The increase in ammonia emissions have made it, along with sulphur dioxide, nitrogen oxides and tropospheric ozone, one of the most worrying pollutants.

3. Data And Methodology

The ammonia emissions (in kilotons) datasets have been obtained from Feng et al. (2020). The collection and processing of the necessary data in a consistent format are done in the first stage of the data preparation. Thereafter, emission factor information gathered in the first stage are utilised to compute default emissions data. The datasets are for 1770–2019.

Dealing with the methodology, we use techniques based on fractional integration, which are very useful for the purpose of describing issues such as persistence, and time trends in time series data. A process $\{x_t, t = 0, \pm 1, \ldots\}$ is said to be fractionally integrated or integrated of order d, and represented as I(d), if it can be expressed as:

$$(1 - B)^d x_t = u_t, \qquad t = 1, 2, \ldots, \qquad (1)$$

where B is the backshift operator (i.e., $B_k x_t = x_{t-k}$) and where u_t is integrated of order 0 or I(0) that means that it is second order stationary with a spectral density function that is positive and bounded at all frequencies. Within the I(0) category we have the white noise process but also other processes allowing, for example, some type of weak (ARMA) autocorrelation.

Using a Binomial expansion on the polynomial in B in the left hand side of (1), x_t can be expressed in terms of all its past history, adopting the form of an infinite AR process,

$$x_t = d\, x_{t-1} - \frac{d(d-1)}{2} x_{t-2} + \frac{d(d-1)(d-2)}{6} x_{t-3} - \ldots + u_t,$$

and thus, the differencing parameter d can be taken as a measure of the degree of persistence of the data, since the higher the value of d is, the higher the association between observations is, even if they are far apart in time. The estimation is conducted via Whittle function in the frequency domain (Dahlhaus, 1989) by implementing a very simple version of Robinson's (1994) tests, widely used in recent years in empirical applications of environmental studies (see, e.g., Nikolopoulos et al., 2019; Caporale et al., 2021, Gil-Alana and Sakiru, 2021; etc.).

4. Empirical Results

We look at the following regression model,

$$y_t = \beta_0 + \beta_1 t + x_t, \qquad (1-L)^d x_t = u_t, \qquad t = 1, 2, \ldots \qquad (2)$$

where y_t refers to the observed time series; β_0 and β_1 are the coefficients corresponding respectively to the intercept and a linear time trend, and x_t is supposed to be I(d) where d is another parameter that is also estimated from the data. Dealing with the error term u_t, we assume first that it is a white noise process, and later, we assume (weak) autocorrelation based on Bloomfield (1973)[1]. Tables 1 and 2 refer to the case of white noise errors, while Tables 3 and 4 to the model of Bloomfield (1973) for the error term.Table 1 shows the values of the differencing parameter, d, and their 95% confidence bands under the three classical assumptions in the unit root literature of: i) no deterministic terms, ii) an intercept and iii) an intercept with a linear time trend, with the selected model for each series presented in bold in the table. The first thing we observe in this table is that the time trend is required in a number of cases, in particular in 13 out of the 37 countries examined; in another group of 22 countries, the intercept is statistically significant, while for two countries (Finland and the USA) both coefficients (intercept and time trend) are found to be statistically insignificant. The estimated coefficients are displayed in Table 2, and the highest time trend coefficient corresponds to Mexico (3.0297), followed by Turkey (2.5666) and Australia (2.1189). Moving now to the estimated orders of integration, we observe that the results are very heterogeneous across the countries: Finland is the only country showing statistical evidence of mean reversion (d < 1); the unit root null (d = 1) cannot be rejected in the cases of Norway or Iceland; for all the other countries the orders of integration are substantially higher than 1.

Table 1
Estimates of d: White noise errors

Country	No terms	An intercept	An intercept and a linear time trend
AUSTRALIA	1.14 (1.07, 1.23)	1.14 (1.07, 1.23)	**1.15 (1.07, 1.24)**
AUSTRIA	1.15 (1.09, 1.23)	1.23 (1.17, 1.30)	**1.23 (1.18, 1.30)**
BELGIUM	1.14 (1.07, 1.22)	**1.15 (1.08, 1.23)**	1.15 (1.08, 1.23)
CANADA	1.19 (1.13, 1.27)	1.18 (1.12, 1.27)	**1.19 (1.13, 1.28)**
CHILE	1.18 (1.13, 1.24)	1.18 (1.13, 1.24)	**1.19 (1.14, 1.25)**
COLOMBIA	1.18 (1.14, 1.24)	1.18 (1.14, 1.24)	**1.19 (1.15, 1.25)**
CZECH REPUBLIC	1.30 (1.22, 1.39)	**1.38 (1.30, 1.46)**	1.38 (1.30, 1.46)
DENMARK	1.36 (1.29, 1.44)	**1.38 (1.31, 1.46)**	1.38 (1.31, 1.45)
ESTONIA	1.46 (1.38, 1.55)	**1.49 (1.41, 1.59)**	1.49 (1.41, 1.59)
FINLAND	**0.59 (0.52, 0.68)**	0.59 (0.53, 0.68)	0.59 (0.52, 0.68)
FRANCE	1.10 (1.04, 1.18)	**1.25 (1.20, 1.31)**	1.25 (1.20, 1.31)
GERMANY	1.32 (1.24, 1.43)	**1.40 (1.31, 1.50)**	1.40 (1.31, 1.50)
GREECE	1.17 (1.11, 1.25)	**1.20 (1.15, 1.27)**	1.20 (1.15, 1.27)
HUNGARY	1.37 (1.28, 1.49)	**1.40 (1.30, 1.52)**	1.40 (1.30, 1.52)
ICELAND	1.05 (0.98, 1.14)	1.05 (0.98, 1.14)	**1.05 (0.98, 1.15)**
IRELAND	1.16 (1.08, 1.25)	**1.33 (1.26, 1.42)**	1.33 (1.26, 1.42)
ISRAEL	1.21 (1.14, 1.31)	**1.22 (1.15, 1.32)**	1.23 (1.16, 1.32)
ITALY	1.06 (0.99, 1.14)	1.10 (1.05, 1.17)	**1.11 (1.05, 1.18)**
JAPAN	1.13 (1.07, 1.22)	**1.22 (1.16, 1.30)**	1.22 (1.16, 1.30)
KOREA	1.24 (1.19, 1.31)	**1.25 (1.19, 1.31)**	1.25 (1.19, 1.31)
LATVIA	1.48 (1.37, 1.64)	**1.72 (1.55, 1.94)**	1.72 (1.55, 1.94)
LITHUANIA	1.42 (1.32, 1.55)	**1.46 (1.36, 1.60)**	1.46 (1.36, 1.60)
LUXEMBOURG	1.22 (1.15, 1.31)	**1.24 (1.17, 1.33)**	1.24 (1.17, 1.33)
MEXICO	1.20 (1.15, 1.25)	1.20 (1.15, 1.25)	**1.21 (1.16, 1.26)**
NETHERLANDS	1.24 (1.18, 1.30)	**1.24 (1.18, 1.30)**	1.24 (1.18, 1.30)

Values in parenthesis indicate the 95% confidence interval of the non-rejection values of d using Robinson (1994). In bold, the selected specification for the deterministic terms in each series.

Country	No terms	An intercept	An intercept and a linear time trend
NEW ZEALAND	1.14 (1.08, 1.21)	1.14 (1.08, 1.21)	**1.15 (1.09, 1.22)**
NORWAY	1.01 (0.95, 1.08)	1.01 (0.95, 1.09)	**1.01 (0.95, 1.09)**
POLAND	1.33 (1.24, 1.43)	**1.34 (1.25, 1.45)**	1.34 (1.26, 1.45)
PORTUGAL	1.12 (1.05, 1.21)	**1.14 (1.08, 1.23)**	1.15 (1.08, 1.23)
SLOVAKIA	1.15 (1.08, 1.23)	**1.15 (1.09, 1.24)**	1.16 (1.09, 1.24)
SLOVENIA	1.07 (1.02, 1.13)	1.08 (1.03, 1.14)	**1.08 (1.03, 1.15)**
SPAIN	1.14 (1.08, 1.21)	1.15 (1.09, 1.22)	**1.15 (1.09, 1.22)**
SWEDEN	1.32 (1.25, 1.41)	**1.39 (1.32, 1.47)**	1.39 (1.32, 1.47)
SWITZERLAND	1.18 (1.11, 1.26)	**1.26 (1.20, 1.31)**	1.26 (1.20, 1.31)
TURKEY	1.15 (1.08, 1.25)	1.16 (1.09, 1.27)	**1.17 (1.09, 1.28)**
UK	1.20 (1.13, 1.29)	**1.23 (1.16, 1.31)**	1.23 (1.17, 1.32)
USA	**1.31 (1.21, 1.43)**	1.31 (1.21, 1.43)	1.31 (1.22, 1.43)

Values in parenthesis indicate the 95% confidence interval of the non-rejection values of d using Robinson (1994). In bold, the selected specification for the deterministic terms in each series.

Table 2
Estimated coefficients in Table 1: White noise errors

Country	d	Intercept (t-value)	Time trend (t-value)
AUSTRALIA	1.15 (1.07, 1.24)	-1.3130 (-0.18)	2.1189 (2.21)
AUSTRIA	1.23 (1.18, 1.30)	9.6768 (16.84)	0.2017 (1.73)
BELGIUM	1.15 (1.08, 1.23)	12.1843 (4.68)	—
CANADA	1.19 (1.13, 1.28)	0.6221 (0.12)	1.6801 (1.97)
CHILE	1.19 (1.14, 1.25)	2.3065 (1.13)	0.7652 (2.29)
COLOMBIA	1.19 (1.15, 1.25)	6.2679 (1.68)	1.5465 (2.53)
CZECH REPUBLIC	1.38 (1.30, 1.46)	21.9430 (11.23)	—
DENMARK	1.38 (1.31, 1.46)	7.7977 (5.93)	—
ESTONIA	1.49 (1.41, 1.59)	1.8773 (5.97)	—
FINLAND	0.59 (0.52, 0.68)	—	—
FRANCE	1.25 (1.20, 1.31)	146.7798 (28.79)	—
GERMANY	1.40 (1.31, 1.50)	77.3950 (9.48)	—
GREECE	1.20 (1.15, 1.27)	6.4508 (6.96)	—
HUNGARY	1.40 (1.30, 1.52)	16.2086 (5.38)	—
ICELAND	1.05 (0.98, 1.15)	0.1397 (1.26)	0.0192 (2.19)
IRELAND	1.33 (1.26, 1.42)	35.3033 (27.48)	—
ISRAEL	1.22 (1.15, 1.32)	2.0047 (5.56)	—
ITALY	1.11 (1.05, 1.18)	65.0932 (11.41)	1.0835 (1.75)
JAPAN	1.22 (1.16, 1.30)	113.0488 (15.56)	—
KOREA	1.25 (1.19 1.31)	9.4460 (2.78)	—
LATVIA	1.72 (1.55, 1.94)	6.1218 (2.83)	—
LITHUANIA	1.46 (1.36, 1.60)	6.7481 (5.90)	—
LUXEMBOURG	1.24 (1.17, 1.33)	0.5513 (7.98)	—
MEXICO	1.21 (1.16, 1.26)	22.7619 (2.89)	3.0297 (2.11)

The values in parenthesis in column 2 are the 95% confidence intervals. In columns 3 and 4 they are t-values.

Country	d	Intercept (t-value)	Time trend (t-value)
NETHERLANDS	1.24 (1.18, 1.30)	10.2021 (1.66)	—
NEW ZEALAND	1.15 (1.09, 1.22)	0.9234 (0.46)	0.7229 (2.73)
NORWAY	1.01 (0.95, 1.09)	1.8700 (3.14)	0.1188 (3.10)
POLAND	1.34 (1.25, 1.45)	39.4288 (4.22)	—
PORTUGAL	1.14 (1.08, 1.23)	7.4941 (6.95)	—
SLOVAKIA	1.15 (1.09, 1.24)	5.4500 (3.06)	—
SLOVENIA	1.08 (1.03, 1.15)	1.6327 (4.65)	0.0597 (1.84)
SPAIN	1.15 (1.09, 1.22)	39.7281 (5.47)	1.7364 (1.79)
SWEDEN	1.39 (1.32, 1.47)	6.3292 (11.17)	—
SWITZERLAND	1.26 (1.20, 1.31)	10.6917 (14.92)	—
TURKEY	1.17 (1.09, 1.28)	52.6523 (5.85)	2.5666 (1.92)
UK	1.23 (1.16, 1.31)	26.7184 (9.02)	—
USA	1.31 (1.21, 1.43)	—	—

The values in parenthesis in column 2 are the 95% confidence intervals. In columns 3 and 4 they are t-values.

Table 3
Estimates of d: Autocorrelated (Bloomfield) errors

Country	No terms	An intercept	An intercept and a linear time trend
AUSTRALIA	1.12 (1.02, 1.28)	1.12 (1.02, 1.28)	**1.14 (1.03, 1.29)**
AUSTRIA	1.32 (1.17, 1.49)	**1.48 (1.32, 1.69)**	1.49 (1.34, 1.69)
BELGIUM	1.22 (1.09, 1.41)	**1.25 (1.12, 1.42)**	1.25 (1.13, 1.42)
CANADA	1.15 (1.08, 1.25)	1.15 (1.08, 1.25)	**1.16 (1.09, 1.27)**
CHILE	**1.29 (1.20, 1.45)**	1.29 (1.20, 1.42)	1.30 (1.21, 1.45)
COLOMBIA	1.27 (1.21, 1.35)	**1.27 (1.21, 1.35)**	1.29 (1.23, 1.37)
CZECH REPUBLIC	1.29 (1.16, 1.47)	**1.41 (1.27, 1.58)**	1.41 (1.27, 1.58)
DENMARK	1.46 (1.33, 1.60)	**1.44 (1.34, 1.59)**	1.44 (1.34, 1.59)
ESTONIA	1.57 (1.37, 1.82)	**1.59 (1.38, 1.81)**	1.59 (1.38, 1.81)
FINLAND	**0.61 (0.50, 0.73)**	0.61 (0.51, 0.73)	0.61 (0.51, 0.73)
FRANCE	1.21 (1.09, 1.36)	**1.64 (1.50, 1.78)**	1.64 (1.50, 1.78)
GERMANY	1.29 (1.14, 1.47)	**1.34 (1.21, 1.55)**	1.34 (1.21, 1.55)
GREECE	1.29 (1.18, 1.33)	**1.36 (1.26, 1.49)**	1.36 (1.26, 1.49)
HUNGARY	1.19 (1.04, 1.38)	**1.18 (1.04, 1.36)**	1.18 (1.04, 1.36)
ICELAND	0.98 (0.90, 1.06)	0.98 (0.91, 1.07)	**0.98 (0.90, 1.07)**
IRELAND	1.23 (1.11, 1.40)	**1.29 (1.17, 1.44)**	1.29 (1.17, 1.44)
ISRAEL	1.22 (1.12, 1.38)	**1.23 (1.12, 1.39)**	1.23 (1.12, 1.39)
ITALY	1.14 (1.03, 1.29)	**1.23 (1.14, 1.36)**	1.24 (1.14, 1.36)
JAPAN	0.88 (0.82, 0.97)	**1.23 (1.05, 1.73)**	1.22 (1.05, 1.73)
KOREA	1.43 (1.32, 1.58)	**1.44 (1.33, 1.58)**	1.44 (1.33, 1.58)
LATVIA	1.12 (0.96, 1.31)	**1.00 (0.87, 1.18)**	1.00 (0.87, 1.18)
LITHUANIA	1.16 (1.00, 1.35)	**1.16 (1.01, 1.34)**	1.16 (1.01, 1.34)
LUXEMBOURG	1.28 (1.14, 1.47)	**1.30 (1.17, 1.47)**	1.31 (1.17, 1.47)
MEXICO	1.38 (1.30, 1.51)	**1.40 (1.31, 1.52)**	1.40 (1.32, 1.52)
NETHERLANDS	1.53 (1.40, 1.69)	**1.52 (1.40, 1.69)**	1.52 (1.40, 1.69)

Values in parenthesis indicate the 95% confidence interval of the non-rejection values of d using Robinson (1994). In bold, the selected specification for the deterministic terms in each series.

Country	No terms	An intercept	An intercept and a linear time trend
NEW ZEALAND	1.20 (1.13, 1.34)	1.20 (1.13, 1.34)	**1.24 (1.14, 1.34)**
NORWAY	1.03 (0.96, 1.15)	1.04 (0.97, 1.17)	**1.05 (0.97, 1.17)**
POLAND	1.21 (1.07, 1.39)	**1.20 (1.06, 1.37)**	1.20 (1.06, 1.37)
PORTUGAL	1.13 (1.03, 1.27)	**1.15 (1.05, 1.28)**	1.15 (1.05, 1.28)
SLOVAKIA	1.28 (1.14, 1.46)	**1.30 (1.16, 1.48)**	1.30 (1.16, 1.48)
SLOVENIA	1.31 (1.19, 1.46)	**1.35 (1.23, 1.50)**	1.35 (1.23, 1.50)
SPAIN	1.59 (1.33, 2.00)	**1.55 (1.31, 2.01)**	1.55 (1.31, 2.01)
SWEDEN	1.37 (1.24, 1.53)	**1.43 (1.31, 1.60)**	1.43 (1.31, 1.60)
SWITZERLAND	1.29 (1.16, 1.47)	**1.41 (1.27, 1.58)**	1.41 (1.27, 1.58)
TURKEY	1.04 (0.97, 1.13)	1.01 (0.95, 1.11)	**1.02 (0.94, 1.11)**
UK	1.27 (1.13, 1.45)	**1.29 (1.17, 1.44)**	1.30 (1.17, 1.44)
USA	1.19 (1.08, 1.38)	1.19 (1.07, 1.38)	**1.19 (1.07, 1.37)**

Values in parenthesis indicate the 95% confidence interval of the non-rejection values of d using Robinson (1994). In bold, the selected specification for the deterministic terms in each series.

Table 4
Estimated coefficients in Table 3: Autocorrelation (Bloomfield) errors

Country	**d**	**Intercept (t-value)**	**Time trend (t-value)**
AUSTRALIA	1.14 (1.03, 1.29)	-1.3425 (-0.18)	2.1273 (2.34)
AUSTRIA	1.48 (1.32, 1.69)	9.7866 (19.12)	—
BELGIUM	1.25 (1.12, 1.42)	12.1852 (4.80)	—
CANADA	1.16 (1.09, 1.27)	0.5516 (0.10)	1.7160 (2.34)
CHILE	1.29 (1.20, 1.45)	—	—
COLOMBIA	1.27 (1.21, 1.35)	7.1340 (2.01)	—
CZECH REPUBLIC	1.41 (1.27, 1.58)	21.9434 (11.35)	—
DENMARK	1.44 (1.34, 1.59)	7.7987 (6.07)	—
ESTONIA	1.59 (1.38, 1.81)	1.8773 (1.59)	—
FINLAND	0.61 (0.50, 0.73)	—	—
FRANCE	1.64 (1.50, 1.78)	146.8149 (35.24)	—
GERMANY	1.34 (1.21, 1.55)	77.3850 (9.30)	—
GREECE	1.36 (1.26, 1.49)	6.4544 (7.39)	—
HUNGARY	1.18 (1.04, 1.36)	16.2063 (5.13)	—
ICELAND	0.98 (0.90, 1.07)	0.1340 (1.21)	0.0195 (3.20)
IRELAND	1.29 (1.17, 1.44)	35.2988 (27.18)	—
ISRAEL	1.23 (1.12, 1.39)	2.0047 (5.57)	—
ITALY	1.23 (1.14, 1.36)	65.7741 (12.01)	—
JAPAN	1.23 (1.05, 1.73)	113.0952 (7.00)	
KOREA	1.44 (1.33, 1.58)	9.4576 (3.02)	—
LATVIA	1.00 (0.87, 1.18)	6.1337 (11.89)	—
LITHUANIA	1.16 (1.01, 1.34)	6.7463 (5.54)	—
LUXEMBOURG	1.30 (1.17, 1.47)	0.5514 (8.15)	—
MEXICO	1.40 (1.31, 1.52)	21.4450 (3.43)	—

The values in parenthesis in column 2 are the 95% confidence intervals. In columns 3 and 4 they are t-values.

Country	d	Intercept (t-value)	Time trend (t-value)
NETHERLANDS	1.52 (1.40, 1.69)	10.2134 (1.89)	
NEW ZEALAND	1.24 (1.14, 1.34)	0.9888 (0.51)	0.6893 (1.68)
NORWAY	1.05 (0.97, 1.17)	1.8860 (3.17)	0.1180 (2.51)
POLAND	1.20 (1.06, 1.37)	39.4106 (4.07)	—
PORTUGAL	1.15 (1.05, 1.28)	7.4941 (6.95)	—
SLOVAKIA	1.30 (1.16, 1.48)	5.4509 (3.20)	—
SLOVENIA	1.35 (1.23, 1.50)	1.6747 (5.28)	—
SPAIN	1.55 (1.31, 2.01)	40.7860 (6.64)	—
SWEDEN	1.43 (1.31, 1.60)	6.3294 (11.38)	—
SWITZERLAND	1.41 (1.27, 1.58)	10.6393 (15.46)	—
TURKEY	1.02 (0.94, 1.11)	51.8872 (5,75))	2.5485 (4.16)
UK	1.29 (1.17, 1.44)	26.7275 (9.22)	—
USA	1.19 (1.07, 1.37)	3.2966 (0.09)	12.6060 (2.11)

The values in parenthesis in column 2 are the 95% confidence intervals. In columns 3 and 4 they are t-values.

Tables 3 and 4 are similar to Tables 1 and 2 but assuming that the error term is autocorrelated. However, instead of imposing a specific ARMA model for the error term, we employ a non-parametric approximation based on Bloomfield (1973). Starting with the results displayed in Table 3, we observe that the time trend coefficient is now significant in only 7 countries (of which 6, the time trend was also significant under white noise errors); for 28 countries the intercept seems to be sufficient, and for Chile and Finland, no deterministic terms are required. Focussing on the estimates of d, we observe that once more, Finland is the only country displaying mean reversion; also, apart from Norway and Iceland, the unit root null rejected cannot be rejected now in the cases of Latvia and Turkey, and the null hypothesis of I(1) is rejected in all the remaining countries in favour of $d > 1$.

Finally, Tables 5 and 6 display summary results in relation with the time trends (Table 5) and with the orders of integration (Table 6). Starting with the time trends, we observe that if u_t is autocorrelated the coefficient for the time trend is very large in the case of the US (12.6060) followed by Turkey, Australia and Canada which also display large positive values under both types of specifications for the error term. These coefficients are all positive, which is not good for the environment. On the other hand, there are 22 countries with insignificant time trends. Looking, finally, at the orders of integration, the results are also robust across the errors, and mean reversion only seems to happen in the case of Finland (0.59 with white noise errors and 0.61 under autocorrelation); Norway and Iceland show evidence of I(1) behaviour under the two specifications and also Latvia and Turkey with Bloomfield disturbances. In the remaining countries, the degree of differentiation is significantly higher than 1.

Table 5
Summary results: Statistical significant time trend coefficients

White noise errors	Autocorrelated errors
MEXICO (3.0297)	USA (12.6060)
TURKEY (2.5666)	TURKEY (2.5485)
AUSTRALIA (2.1189)	AUSTRALIA (2.1273)
SPAIN (1.7364)	CANADA (1.7160)
CANADA (1.6801)	NEW ZEALAND (0.6893)
COLOMBIA (1.5465)	NORWAY (0.1180)
ITALY (1.0835)	ICELAND (0.0195)
CHILE (0.7652)	
NEW ZEALAND (0.7229)	
AUSTRIA (0.2017)	
NORWAY (0.1188)	
SLOVENIA (0.0597)	
ICELAND (0.0192)	

Table 6
Summary results: Orders of integration

White noise errors			Autocorrelated errors		
d < 1	d = 1	d > 1	d < 1	d = 1	d > 1
FINLAND (0.59)	NORWAY (1.01)	SLOVENIA (1.08)	FINLAND (0.61)	ICELAND (0.98)	AUSTRALIA (1.14)
	ICELAND (1.05)	ITALY (1.11)		LATVIA (1.00)	PORTUGAL (1.15)
		PORTUGAL (1.14)		TURKEY (1.02)	LITIHUANIA (1.16)
		NEW ZEALAND (1.15)		NORWAY (1.05)	C.ANADA (1.16)
		SLOVAKIA (1,.15)			HUNGARY (1.18)
		SPAIN (1.15)			POLAND (1.20)
		TURKEY (1.17)			ISRAEL (1.23)
		CANADA (1.19)			ITALY (1.23)
		CHILE (1.19)			JAPAN (1.23)
		COLOMBIA (1.19)			NEW ZEALAND (1.24)
		GREECE (1.20)			BELGIUM (1.25)
		MEXICO (1.21)			COLOMBIA (1.27)
		ISRAEL (1.22)			IRELAND (1.29)
		JAPAN (1.22)			CHILE (1.29)
		UK (1.23)			UK (1.29)
		NETHERLANDS (1.24)			SLOVAKIA (1.30)
		FRANCE (1.25)			LUXEMBOURG (1.31)
		KOREA (1.25)			GERMANY (1.34)
		SWITZERLAND (1.26)			SLOVENIA (1.35)
		USA (1.31)			GREECE (1.36)
		IRELAND (1.33)			MEXICO (140)
		POLAND (1.34)			SWITZERLAND (1.41)
		CZECH REP. (1.38)			CZECH REP. (1.41)
		DENMARK (1.38)			SWEDEN (1.43)

White noise errors		Autocorrelated errors	
	SWEDEN (1.39)		KOREA (1.44)
	GERMANY (1.40)		DENMARK (1.44)
	HUNGARY (1.40)		AUSTRIA (1.48)
	LITUANIA (1.46)		NETHERLANDS (1.52)
	ESTONIA (1.49)		SPAIN (1.55)
	LATVIA (1.72)		ESTONIA (1.59)
			FRANCE (1.64)

One of the justifications for the foregoing empirical findings is that the drivers of ammonia tend to be persistent. According to Narayan (2007), a series which is dependent on other series which are persistent will inherit this persistence, and transmit to several other series in a country. Nguyen et al. (2020) has shown that determinants of ammonia emissions- income per capita, energy consumption per capita and foreign direct investment are very persistent.

5. Concluding Comments

We have investigated in this work the statistical properties of ammonia (NH_3) historical time series data in 37 countries for the time period from 1770 to 2019, annually. Using fractional integration methods our results indicate that reversion to the mean only takes place in the case of Finland, while the unit root hypothesis cannot be rejected for Norway or Iceland. In the remaining cases, the estimated values of d are much higher than 1, and this result is robust across the different specifications for the error term.

An implication of the empirical results of this study is that, shocks to ammonia emissions in these countries will have permanent effects. Therefore, a combination of appropriate policies and technologies should be adopted to address any upsurge in ammonia emissions. There are several policies that can be utilised to address ammonia emissions such as the introduction of emission tax, a total ban on solid urea fertilisers, the funding and expansion of conservation areas, offering incentives to assist suppliers of sustainable commodities, improving private sector participation in the supply chains of agricultural products.

The available technologies include condensers (which are utilised to eradicate ammonia by converting the gas to a liquid), wet scrubbers (which are devices used in removing ammonia from furnace flue gas or from other gas streams), urease inhibitor (which is a chemical that assists the slowing down of the conversion of urea to ammonium) and the recycling of ammonia. Countries such as the UK are in the process of introducing large scale solid urea fertilisers (Society of Chemical Industry, 2020)

Other modelling approaches still within the context of fractional integration can be taken into account. Thus, for example, non-linearities and breaks are topics which are likely to occur when using long historical data, and many authors have found that this I(d) specification is very much related to these two issues (Diebold and Inoue, 2001; Granger and Hyung, 2004; Ohanissian et al., 2008; etc.). Then, alternative non-linear deterministic approaches, based, for example, on Chebyshev's polynomials in time (Cuestas and Gil-Alana, 2016) or on Fourier transforms (Yaya et al., 2020) can be used in these or in alternative datasets.

Declarations

Ethical Approval

Not applicable.

Consent to Participate

Not applicable.

Consent to Publish

Not applicable.

Authors Contributions

Prof. Solarin Sakiru obtained the data. He worked on the introduction and literature review. He also contributed with the empirical results and conclusions.

Prof. Lorenzo Bermejo made part of the introduction and the literature review along with the conclusions.

Prof. Luis A. Gil-Alana proposed the original idea; he conducted the programming and the empirical results along with their interpretation.

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Competing Interests

The authors declare that they have no competing interests.

Availability of data and materials

Data are availbale from the authors upon request.

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